Schwoebel Effect and Dynamic Scaling Behavior in Nickel Film Growth by Electrodeposition

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Two kinds of the growth rate of nickel films by electrodeposition were measured by use of columnar photo-resists formed on ITO glass plates. The ratio of the mean growth rate at the edge to that of the nickel layer was 1.5, which indicates direct evidence for the Schwoebel effect [R. L. Schwoebel, J. Appl. Phys. 40,614(1969)], i.e., the presence of the anisotropy between the up and down step incorporation probabilities of adatoms. The growth exponent, 0.3 was obtained from the AFM images of the nickel film surface. This suggests that the nickel growth by electrodeposition has a dynamic scaling property.

Keywords: nickel electrodeposition, Schwoebel effect, growth exponent, AFM, dynamic scaling

1. Introduction

Many studies on the morphology and the dynamics of growth in thin solid films have been made to understand the phenomena of growth by electrodeposition [1]. As is well known, a large number of factors are found to influence the formation of the surfaces [2,3]. Nevertheless, it is desirable to describe the growth mechanism by a few basic factors.

For example, Schwoebel [4] found the difference in the up and down step incorporation probabilities of deposited atoms that determines the surface morphology. If the probability captured at the up step is less than that at the down step, the step between its neighbors becomes wider in time. In this study, in order to make clear the presence of the capture anisotropy, two kinds of the growth rate in nickel film electrodeposition, i.e., the growth rate at the edge (see Fig.1) and that of the nickel film were measured by use of columnar photo-resists formed on ITO (indium tin oxide) glass plates.

On the other hand, lately the dynamic scaling theory [5] has made clear small numbers of the exponents determining the morphology and the dynamics of growth. These exponents are related to the roughness of the thin film surfaces [5,6]. If thin film growth by electrodeposition has a scaling property, the surface roughness of the film W increases in time as $W \sim t^{\beta}$ and saturates at a size of the nickel film as $W_{sat} \sim L^{\alpha}$ where α and β are called the roughness exponent and the growth exponent, and L is a system size. The experimental values of the exponents in a dry process such as MBE, sputtering and ion bombardment experiments have already been reported [5]. We have interests in the dynamic scaling laws of growth in a wet process such as electrodeposition. However very few experiments on the exponents in electrodeposition have been made [7]. In this study a nickel electrodeposition process rather than electrodeposition of alloys was selected owing to its simplicity.

This paper aims at direct evidence for the anisotropy of probabilities of adatoms captured at the edge and its neighbor steps in nickel electrodeposition and the exponents obtained from the roughness of the nickel film surface using AFM (atomic force microscope) [8].

2. Experiment

First, ITO glass plates of $2x4cm^2$ (sheet resistivity $6\Omega/square$) were prepared for cathode plates. A pattern was formed on the ITO glass covered with a photoresist using a lithographic process. The photoresist used was a positive one for

electrodeposition. The pattern comprises of columns of diameter 1mm and thickness 1.6 μ m as shown in Fig.1. The columns become the origin of pores formed in a nickel film during electrodeposition. According to the Schwoebel theory [4], if there is no difference in the probabilities of nickel adatoms captured at the edge and its neighbor steps, the average increase rate of the nickel film height is equal to the decrease rate of the pore diameter. Thus by measuring the two kinds of the growth rates, we can directly confirm the Schwoebel effect. The ITO glass plate was located in a bath containing (g/l): nickel sulfamate, 600; nickel chloride, 5; and boric acid, 40. A nickel plate that has the same dimension as the ITO glass plate was placed parallel in front of the ITO glass at a distance of 4cm. This is because a uniform current distribution is considered to be important to obtain a uniform thin film. The bath was maintained at pH 4, the temperature of 323 K and a constant voltage of 4 V. The diameter of the pore like a tube, D_{Ni} in Fig.1 was measured by a laser profile micrometer and microscope (Keyence VF-7510) to determine the growth rate of the nickel films at the edge.

3. Results and Discussion

The surface roughness of the ITO glass plate was within a range of about 50 nm. All nickel layers deposited on the ITO glass plates reflected light just as a mirror. Fig.2 shows the photograph of the nickel layer grown on the ITO glass plate for 300 sec. The circles on the nickel surface shown in Fig.2 are pores.

The height of the surface at many points was first measured to evaluate the growth rate of the deposited nickel films. Fig.3 shows the dependence of the mean height on deposition time. It's obvious that the mean growth rate of the nickel film holds constant during electrodeposition. This indicates that the number of nickel adatoms arriving on the surface was constant. The mean growth rate of the nickel film in this study was as follows,

$$V_v = 1.52 \times 10^{-2} \ \mu m/sec.$$
 (1)

Second, the changes of the diameter of the pores with electrodeposition time were measure by a laser profile micrometer and microscope. Fig.4 shows a decrease in the diameter of the pore with deposition time. The best straight line fitting to Fig.4 becomes

$$(D_{Re} - D_{Ni})/D_{Re} = 4.69 \times 10^{-5} t - 5.13 \times 10^{-3}$$
, for $t \ge 1.09 \times 10^{2}$. (2)

where D_{Re} is a diameter of the resist column, in this study, 1mm and D_{Ni} a diameter of the pore as shown in Fig.1. The time t= 1.09×10^2 sec is one necessary for the nickel film on the ITO to grow the height of the resist height $1.6\mu m$. The ratio of the growth rate at the edge V_E to the growth rate of the nickel film V_v is obtained from equations (1) and (2),

 $V_{\rm E}/V_{\rm v}=1.5.$ (3)

According to the Schwoebel theory, this result shows the presence of the anisotropy between the edge and its neighbor step incorporation probabilities. If there is no difference in the capture probabilities, we will have $V_{E}\approx V_{v}$. However we have no idea as to why the probability captured at the edge is larger than that at its neighbor steps.

Next, we examined a scaling property of the nickel film deposition. The growth exponent $\boldsymbol{\beta}$ is defined by

 $W(\mathbf{x},\mathbf{t}) \sim \mathbf{t}^{\beta} \tag{4}$

where the surface roughness $W(\mathbf{x},t)=(\Sigma(h(\mathbf{x},t) - h)^2/N)^{1/2}$ where $h(\mathbf{x},t)$ indicates the height of the surface at a position \mathbf{x} and time t, and h is defined by $\Sigma h(\mathbf{x},t)/N$ where N is a number of sites measured. The function $W(\mathbf{x},t)$ and h are called the standard deviation of the roughness and mean height of the surface. To measure the roughness of the nickel layer surface, AFM (Shimazu SPM-9500) images of $10x10 \ \mu m^2$ were taken. Fig.5 shows an AFM image of the nickel layer deposited for 300 sec. Fig.6 shows the logarithmic plot of W (t) and t. As is well known, the random growth model such as a random deposition process with surface relaxation has the growth exponent $\beta=0.5$ [5]. In this study the value of β is found to be 0.3 less than 0.5 in random deposition. This suggests that the surface morphology in nickel electrodeposition has a dynamic scaling property. However we have not found a saturation time of the surface roughness, i.e., a value of the roughness exponent α . It is not clear at present that this is due to the electrodeposition time that was too short to reach a saturated value, the occurrence of instability in the surface predicted by Schwoebel or other reasons.

3. Conclusion

The two kinds of the growth rates in nickel electrodeposition were measured by use of columnar photo-resist formed on ITO glass plates. The ratio of the two mean growth rates 1.5 made clear the presence of the Schwoebel effect. The growth exponent, 0.3 analyzed by the AFM images of the nickel surface suggests the presence of a scaling property in nickel electrodeposition.

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Figure Captions

- Fig.1 Illustration of a pattern formed on the ITO glass plate covered with a photoresist. D_{Re} : diameter of a photoresist column, D_{Ni} : diameter of a pore formed on the resist column.
- Fig.2 Photograph of the nickel layer deposited for 300 sec.
- Fig.3 Dependence of the height of the nickel layer on time.
- Fig.4 Change of $(D_{Re} D_{Ni})$ with electrodeposition time.
- Fig.5 AFM image of the nickel film surface deposited for 300 sec.
- Fig.6 The growth exponent for the nickel layer calculated from the roughness of the nickel film surface using the AFM images.



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Fig.2 Photograph of the nickel layer deposited for 300 sec.



Fig.3 Dependence of the height of the nickel layer on time.



Fig.4 Change of ($D_{\mbox{\scriptsize Re}\mbox{-}}$ $D_{\mbox{\scriptsize Ni}}$) with electrodeposition time.



Fig.5 AFM image of the nickel film surface deposited for 300 sec.



Fig.6 The growth exponent for the nickel layer calculated from the roughness of the nickel film surface using the AFM images.