Fabrication and Characterization of Ni Thin Films Using Direct-Current Magnetron Sputtering *

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Ni films are deposited by using ultra high vacuum dc magnetron sputtering onto silicon substrates at room temperature, and the high-quality and high-density films are prepared. The parameters, such as thickness, density and surface roughness, are obtained by using small-angle x-ray diffraction (XRD) analyses with the Marquardt gradient-expansion algorithm. The deposition rate is calculated and the Ni single layer can be fabricated precisely. Based on the fitting results, we can find that the surface roughness of the Ni films is about 0.7 nm, the densities of Ni films are around 97% and the deposition rate is 0.26 nm/s. The roughness of the surface is also characterized by using an atomic force microscope (AFM). The changing trend of the surface roughness in the simulation of XRD is in good agreement with the AFM measurement.

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Metal–ceramic systems become more interesting in applications and theory. In particular, thin magnetic films containing the $3d$ magnetic atoms Ni, Fe and Co have been thought of as magnetic and magneto-optical storage media.[1] For applications in which the electronic, mechanical and magnetic properties are important, it is also necessary to learn more fundamental aspects of systems such as density and thickness of a film and to know how they are related to the experimental growth conditions. Due to advances in experimental techniques,[2−4] it is now possible to have significant control over the growth conditions of the films and it is, in certain situations, possible to tailor the properties of the films to specific applications. Also, it is important to investigate and to increase the understanding of the structure and growth mechanisms of the thin films. Some experiments[5] depend strongly on accurate knowledge of the film thickness and density of the materials under investigation. In this Letter, the film thickness and density are determined by fitting the small-angle x-ray diffraction data. The Ni films, which are prepared by using the ultra high vacuum dc magnetron sputtering, show good mechanical and magnetic properties, and the densities of the Ni films are close to the bulk density. The films are measured by small-angle x-ray diffraction (XRD) and atomic force microscopy (AFM). Some parameters were given by fitting the measured data, and the deposition rate could be calculated precisely.

Ni films were prepared by ultra high vacuum dc magnetron sputtering from a 10-cm-diameter target of Ni (purity 99.97%) in Ar (purity 99.999%). Before deposition, the sputtering chamber was evacuated to a base pressure of 5×10^{-5} Pa. The sputtering took place at a pressure of 0.26 Pa, a discharge voltage of 290 V and a discharge power of 30 W. The films were deposited onto 20×30 mm silicon substrates, which were placed about 6 cm over the target. The substrate temperature was about room temperature. The deposition time was determined by a stepping motor, which was controlled by a computer.

It is important to investigate and to increase the understanding of some parameters of the thin films. A precise measurement of these parameters can be obtained by the curve fitting of the small-angle x-ray refraction data. The different thickness samples were fabricated by dc magnetron sputtering and then measured by small-angle XRD. The x-ray diffractometer is a D^1 system, which is produced by the Bede Scientific Instruments Ltd. The beam of x-rays from copper K_α emission at wavelength of 0.154 nm was generated and passed through the double silicon channel cut crystal, which can collimate the beam and can filtrate out the copper $K_{\alpha 2}$. For diffraction at small incident angles, the XRD reflectance was obtained by measuring the diffracted beam intensity as a function of the diffraction angle. A series of Ni thin films were fabricated and measured by XRD, as shown in Figs. 1 and 2.

To fit the small-angle XRD data, the so-called Marquardt gradient-expansion algorithm[6] was used. We minimize the value of the statistic S that is defined as

$$S \equiv \sum_{i=1}^{N_m} \frac{(Y[i] - Y_m[i])^2}{w[i]^2}, \qquad (1)$$

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where $w[i]$ are the weighting factors for each point. In conjunction with the nonlinear least-square fitting, some film parameters could be determined by the experimental data.

In order to utilize the nonlinear least-square curve fitting to determine the fitting parameters, the choice of the initial parameter values must be relatively close to the final values, otherwise the fitting algorithm will be unable to locate at the global minimum in parameter space corresponding to the minimum value S. In order to fit the data, one might start by manually varying the Ni single layer thickness, until the fitting reflectance is reasonably coincident with that in the measured data. The measured and fitting curves of grazing-incidence x-ray diffractive intensity versus the grazing angle for a Ni single thin film are shown in Fig. 1. Three adjustable parameters: the density, layer thickness, and interface roughness of the Ni single layer were obtained by fitting the experimental data.

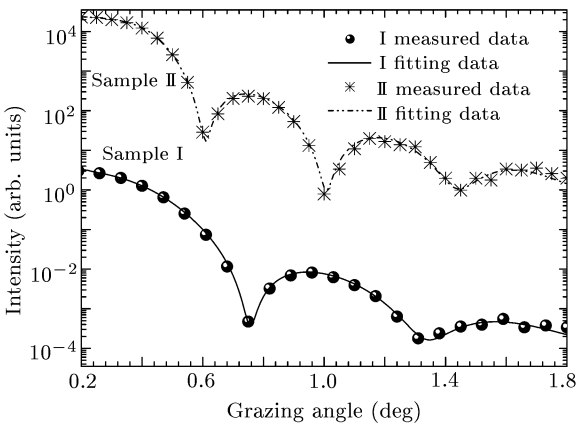


Fig. 1. The measured and fitting curves of x-ray diffractive intensity versus the grazing incidence angle for the Ni single thin film with different deposition time, and samples I and Ⅱ are deposited in time 20 s and 30 s, respectively.

From Fig. 1, we can find that the diffracted curves have different peaks in a certain angular range. The number of diffracted peaks increases with the thickness of the films. The fitting data well match the measured one, and the fitting results are listed in Table 1. Based on the results of samples I and Ⅱ, we find that the thickness is not proportional to the deposition time exactly. In fact, there is not any associated shutter over the target in our magnetron sputtering system, and the deposition time does not include the time that the sample go in and come out the target, which is also the reason why the line does not pass through the origin in Fig. 3. When the films are very thin, the roughness between the substrate and the film also influences the diffracted curves greatly. Therefore, we must increase the deposition time to decrease the fitting error.

In this study, the film thickness and density were determined by fitting the x-ray diffraction data as described before. For a single layer film, the deposition rate can be determined by growing a series of films under the same deposition conditions with different deposition times. By fitting the reflectance data obtained from XRD by the Fresnel equation, the film thickness can be determined. Parameters such as roughness and density were also included in the simulation. The deposition rate can then be determined from the slope of the fitting thickness versus time.

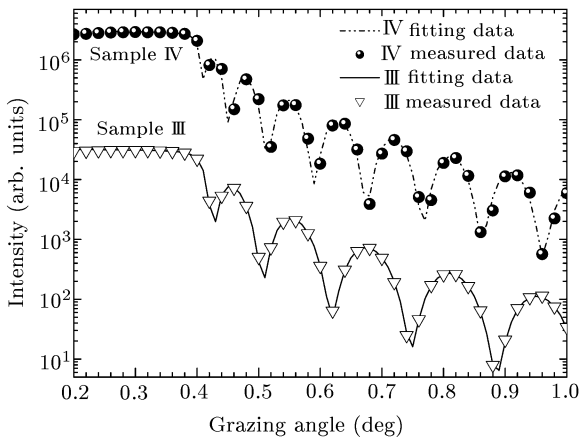


Fig. 2. The measured and fitting curves of x-ray diffractive intensity versus the grazing incidence angle for the Ni single thin film with different deposition time, and samples Ⅲ and Ⅳ are deposited in 100 s and 200 s, respectively.

The measured and fitting curves of x-ray diffractive intensity versus the grazing incidence angle for the Ni single thin film with different deposition time are plotted in Figs. 1 and 2. All the fitting curves coincide with the measured data greatly. The deposition time of samples Ⅲ and Ⅳ is 100 s and 200 s in Fig. 2, and the fitting thickness is 27.9 nm and 55.9 nm, respectively. The data were fitted by using the optical constants taken from Ref. [7]. The surface roughness of the Ni film is expressed in the form of the Debye-Waller factor.[8] Since the densities of the Ni films are close to the buck density by the dc magnetron sputtering and the roughness can be characterized by TEM and AFM. The deposition rate of the Ni thin film can be obtained by using the fitting results. The slope of the fitting thickness versus deposition time yields the deposition rate as illustrated in Fig. 3. The fitting thickness is proportional to the deposition time, which indirectly proves the reliability of the parameter simulation. Based on this rate, any thickness of the Ni film can be fabricated by using the calculated deposition time theoretically. In fact, the deposition rate will exhibit the nonlinearity at very short deposition times, which limits the reliability of the minimum thickness. Based on the above fitting results, we can find that the surface roughness of the Ni films is about 0.7 nm, and the density of film is close to the bulk density. The high-quality and high-density films can be prepared by the dc magnetron sputtering.

Table 1. The fitting results of the samples for different deposition time and the density expressed in the ratio to the bulk density. The roughness is of the film surface.

Sample	Time (s)	Thickness (nm)	Density (%)	Roughness (nm)
Ⅰ	20	6.8	96.57	0.65
Ⅱ	30	9.5	96.95	0.67
Ⅲ	100	27.9	97.88	0.71
Ⅳ	200	55.9	98.00	0.74

In order to verify the reliability of the simulation of roughness, the Ni films are also measured by the AFM (XE-100 SPM System). If the measured surface topography is represented as a surface profile $z(x)$, the rms roughness R_q is defined as the root mean square of the deviations of the surface profile $z(x)$ from the mean line, i.e.

$$R_q = \left[\frac{1}{L}\int_0^L z^2(x)dx\right]^{1/2}, \qquad (2)$$

where L is the length of the surface profile along the x-direction. Accurately measuring surface roughness requires understanding the capabilities and drawbacks of your instrument.[9−11] It is important to believe that the results of a surface measurement with two different types of instruments will agree. This is not easy to demonstrate because different surface-roughness instruments can sample a surface over different spatial frequency ranges.

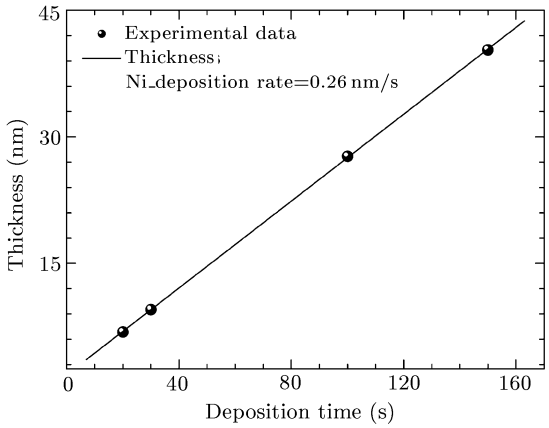


Fig. 3. Deposition rate calculated by fitting the different-deposition-time samples. Here the thickness is the product of the rate and time.

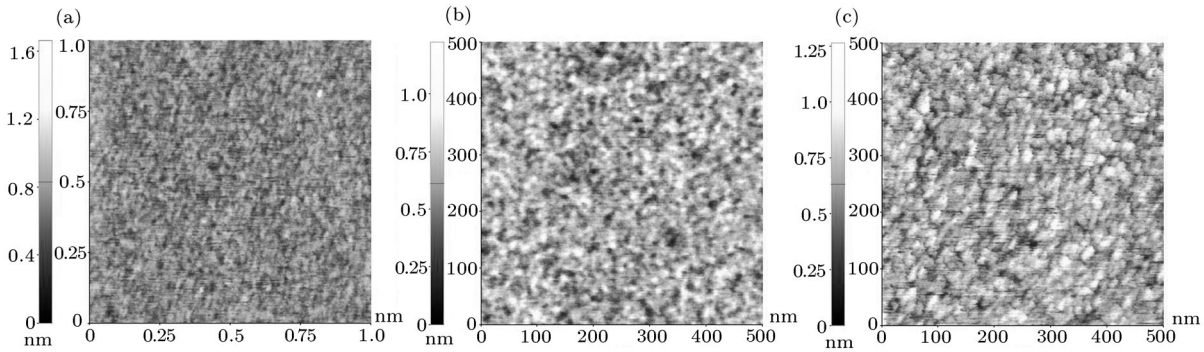


Fig. 4. AFM images of the Ni films with (a) time=30 s, R_q = 0.136 nm, (b) time=100 s, R_q = 0.149 nm, (c) time=150 s, R_q = 0.156 nm.

The scan areas were 0.5 μm×0.5 μm and 1.0 μm×1.0 μm. Surface roughness R_q values were obtained from a complete scan, using the software XEI. For these samples, the scans were repeated from different regions of the sample. In general, this can reveal good reproducibility. Figures 4(a)–4(c) show the AFM topography images of the Ni films with deposition time 30 s and rms roughness 0.136 nm, 100 s and 0.149 nm, 150 s and 0.156 nm, respectively. The roughness of thinner Ni film surface is smaller than the thicker Ni film. This is in agreement with the simulation result of XRD.

In summary, we have carried out the fabrication of the Ni thin film by the dc magnetron sputtering. Some thin films are measured by small-angle XRD and the fitting method is given. The deposition rate of the Ni thin film is calculated precisely. The determination of deposition rate represents a new method for the measurement of thin film thickness, and the experimental data prove its practicability. The surface roughness is characterized by both the small-angle XRD and AFM with the same changing trend. The densities of the Ni films are close to the bulk density and the high density metal films can be deposited by the dc magnetron sputtering method.

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