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Characterization of the surface topography and nano-hardness of Cu/Ni multilayer structures

Research Article

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Abstract: This article describes the results of a study of Cu/Ni multilayer coatings applied on a monocrystalline Si(100) silicon substrate by the deposition magnetron sputtering technique. Composed of 100 bilayers each, the multilayers were differentiated by the Ni sublayer thickness (1.2 to 3 nm), while maintaining the constant Cu sublayer thickness (2 nm). The multilayer coatings were characterized by assessing their surface topography using atomic force microscopy and their mechanical properties with nano-hardness measurements by the Berkovich method. The tests showed that the hardness of multilayers was substantially influenced by the thickness ratio of Cu and Ni sublayers and by surface roughness. The highest hardness and, at the same time, the lowest roughness was exhibited by a multilayer structure with a Cu-to-Ni sublayer thickness ratio of 2 : 1.5.

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Keywords: Cu/Ni multilayer • nanohardness • atomic force microscopy

1. Introduction

Nanomaterials are considered to be particles with one dimension in the size range of 1 nm – 100 nm and that possess novel properties not present in larger particles of the same material. This term can also be applied to multilayers, if the period thickness is within these limits [1]. Multilayers constitute a group of materials with a periodical structure composed of layers of different materials (such as metal, semiconductors, ceramic materials), which may have different physical and mechanical properties. The appropriate selection of layer constituents may cause the resulting coatings to exhibit much better properties compared to monolayers. Multilayers consisting of alternating magnetic and non-magnetic layers are now the subject of studies in the field of materials engineering owing to their high technological potential resulting from their unique magnetic properties, namely the gigantic magnetoresistance effect [2–6].

It has been demonstrated that multilayers consisting of nanometer-order layers exhibit better mechanical properties, which makes them candidates for wear-resistant

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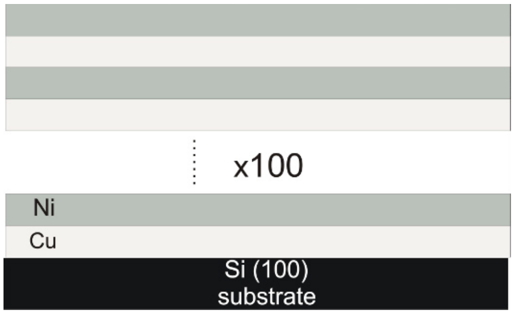

Figure 1. Schematic of the arrangement of sublayers in the multilayers examined.

applications. Many factors contribute to the mechanical properties of multilayers, e.g.: material components, manufacturing process technique and parameters, microstructure and, above all, the quality of the phase boundary [7]. Great progress has been made in recent years in the development of non-destructive and nano-non-destructive techniques used for the characterization of materials at the nano scale. The most important non-destructive methods for the characterization of multilayers are atomic force microscopy (AFM) and X-ray diffraction [8, 9]. The most common method for the determination of the mechanical characteristics of thin-layered structures is the measurement of surface nano-hardness [10].

2. Material and methodology

The material used for tests were Cu/Ni multilayers fabricated by d non-reactive magnetron sputtering on a monocrystalline Si(100) silicon substrate (Fig. 1). The Cu/Ni multilayers were deposited at room temperature. The target materials were copper and nickel (99.99 wt. % purity) 50 mm in diameter. During deposition 99.999% of highly pure Ar gas was used at a pressure of $5 \cdot 10^{-7}$ hPa. The thickness of the layers in the multilayers was controlled by the sputtering time. A deposition speed of 0.126 nm/s was used and the power generated on the magnetron cathode was 0.8 W/cm^2 ($U = 1600$ V, $I = 10$ mA).

The multilayers were differentiated by Ni sublayer thickness, while maintaining a constant thickness of the Cu sublayer (Table 1). Each of the multilayers was built from 100 bilayers, each of a thickness of 3.2, 3.4, 3.5, 3.6, 4.5 and 5.0 nm, respectively. The period thicknesses, being the sum of the thicknesses of two layers, were estimated by X-ray diffractometric examination (XRD) based on the position of the satellite peaks, with an uncertainty within ±0.5 nm [9].

Table 1. The thickness of the sublayers of Cu/Ni multilayers.

Sample no.	1	2	3	4	5	6
Cu sublayer thickness, nm	2	2	2	2	2	2
Ni sublayer thickness, nm	1.2	1.4	1.5	1.6	2.5	3
Total thickness, nm	320	340	350	360	450	500

The multilayer coatings were characterized by examining their surface topography using a Veeco atomic force microscope. The surface scanning area was 1 μm and for each multilayer, five measurements were taken in randomly selected locations. Surface roughness, calculated as the arithmetic mean roughness, R_a, was estimated using the Nanoscope program. Nano-hardness measurements by the Berkovich method were also made using a CSM Nano/Micro-Hardness Tester. The load value was determined experimentally based on the average layer thickness. The load was selected so that the indentation depth equaled approximately 1/4 multilayer thickness and was equal to 1 mN. Five measurements were made on each sample, and the result reported herein represents the arithmetic mean of all measurements. The loading and unloading rate was 2 mN/min.

3. Results

3.1. Multilayer surface topography

Figure 2 presents three-dimensional images of the surface topography of monocrystalline Si(100) constituting the substrate for the multilayers and sample Cu/Ni multilayers, made by the AFM technique. The substrate roughness is approximately two times lower compared to the multilayer roughness. The surfaces of all multilayers are characterized by globular protrusions differing in sizes. The changes in surface topography result from the different period thickness in each multilayer. Based on the average size of globular protrusions, the grain size and the value of the arithmetic mean roughness deviation, R_a, were determined (Fig. 3a). The grain size of the examined multilayers was 25 – 40 nm, and the largest grains were found for multilayers with the Cu/Ni sublayer ratio equal to 2/1.5 and 2/1.6. It was found that the multilayers with the largest grains (40 nm) have the lowest roughness ($R_a = 0.21$ nm).

3.2. Nano-hardness of multilayers

The hardness measurements were made by the Berkovich method. The hardness results were calculated from the

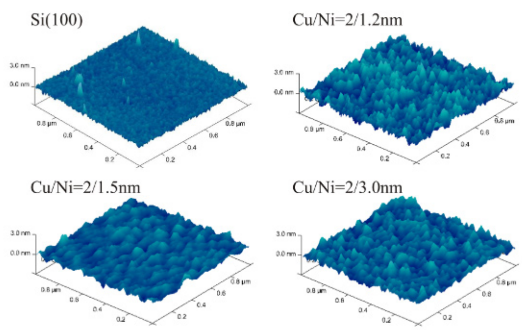

Figure 2. Three-dimensional images of the surface topography of substrate and sample Cu/Ni multilayers subjected to AFM examinations.

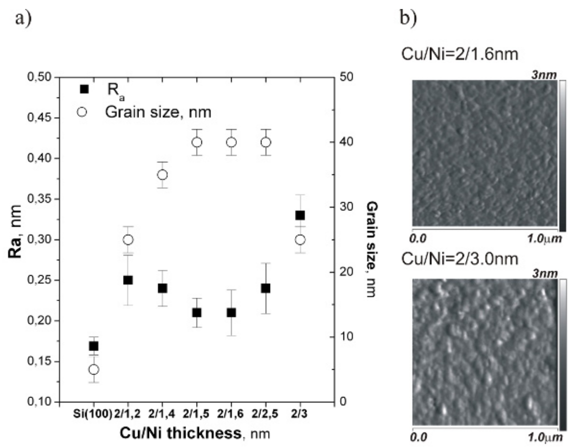

Figure 3. Surface roughness and grain size determined from the area of 1 μm as a function of Cu/Ni multilayer period thickness; b) a two-dimensional image of surface topography for multilayers with the lowest roughness (top/bottom?) and the highest roughness, respectively.

formula:

$$H = \frac{F_{\max}}{A}, \tag{1}$$

where:

$F_{\max}$ — maximum loading force

A — area of contact between the indenter and the multilayer

The curves of loading (up to 1 mN) and unloading of multilayers, as recorded during the measurement, are shown in Figure 4. The behavior of the curves, deviating from a linear response, indicates the elastic reaction of the substrate to the load applied. Based on the recorded penetration depth, the number of bilayers covered directly by the nano-hardness test was estimated. The calculation shows that in the multilayer with the thinnest Ni thickness the measurement encompassed 28 periods (∼ 25%), whereas in the thickest multilayer the measurement covered approx. 17 periods. It should be assumed that the penetrator has a deforming effect on the deeper lying multilayers. The hardness result may also be affected by the substrate; however, all of the multilayers were deposited on monocrystalline silicon, so its effect can be assumed to be similar for each measurement.

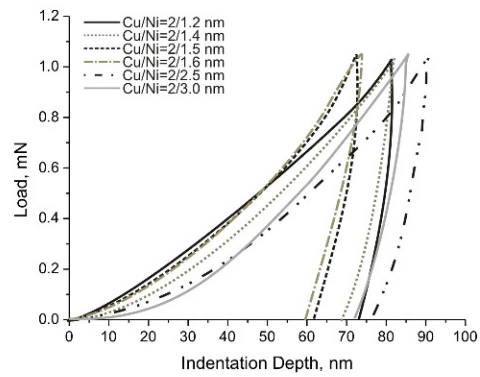

Figure 4. Sample loading and unloading curves for Cu/Ni multilayers

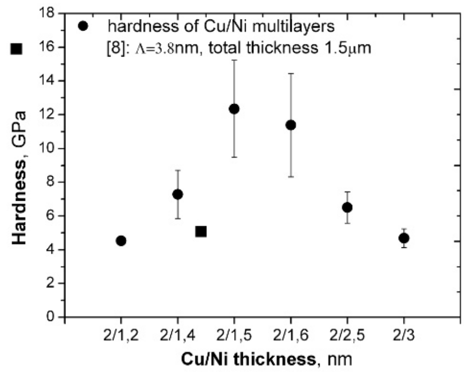

Figure 5. Nano-hardness of the multilayer as a function of Cu/Ni period thickness.

The nano-hardness measurement results (Fig. 5) correlate with the degree of surface development, namely the multilayers with the least roughness have the highest hardness. Therefore, as the measurements of the surface roughness demonstrated a similar trend, it can be suggested that the highest hardness is exhibited by multilayers, in which Ni sublayers are by about 1/4 thinner than the Cu sublayers (Cu/Ni = 2/1.5 and 2/1.6 nm).

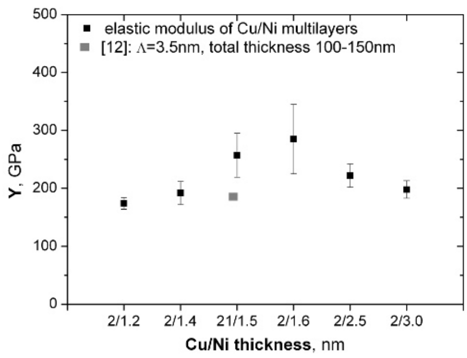

Figure 6. Young's modulus for the multilayer as a function of Cu/Ni period thickness.

The determined multilayer hardness values were compared with values reported by other authors. It was found that the hardness values are close to those presented in reference [7] for the case of a multilayer with similar sublayer thicknesses, 1.9/1.9, but with an overall thickness three times greater (Fig. 5), also fabricated by the magnetron sputtering technique. The microhardness values are also comparable with values reported in reference [11] for similarly composed multilayers deposited electrolytically, with an overall thickness ten times greater.

The Young's modulus (Fig. 6) of the sublayers was determined from the slope of the unloading curve (Fig. 4). The calculated values of Young's modulus are approximately. twice as big as the Young's modulus for solid copper and nickel alloys. The work by X. Y. Zhu *et al.* gives Young's modulus for a Cu/Ni multilayer with a period thickness of 3.5 nm and a total thickness of 100 – 150 nm, obtained using ultra high vacuum technique (UHV). The results provided by the above-mentioned authors are close to those determined in the present work [12].

The research showed that slight differences in the Cu/Ni multilayer period thickness around 0.1 nm caused a change of the hardness of the examined multilayers by several GPa. These changes result, inter alia, from the different moduli of elasticity of the multilayers examined. Higher-hardness multilayers have higher Young's modulus values.

4. Summary

This article has presented the results of AFM and nano-hardness examinations of Cu/Ni multilayers deposited on a monocrystalline silicon substrate. The examined multilayers are characterized by a varying degree of surface development. The characterization of topography, as expressed using the R_a parameter, revealed variations in roughness, depending on the multilayer period thickness, in the range of 0.21 – 0.33 nm, which corresponds to approx. 5% of the bilayer thickness.

Nano-hardness tests showed relatively high multilayer hardness (4.5 – 12.3 GPa) and high elastic modulus (174 – 285 GPa), which predestines the multilayer to applications, in which high wear resistance is required. A correlation has also shown to exist between surface roughness, surface hardness and elastic modulus. The best combination of both of these properties is exhibited by multilayers, in which the Ni sublayer is by approximately 1/4 thinner than the Cu sublayer (Cu/Ni=2/1.5 and 2/1.6 nm).

The occurrence of a relatively large difference in hardness and Young's modulus values while the thickness of one of the multilayers was changed only slightly (about 0.1 nm) suggests, that these values are also influenced by other multilayer features, and not only by roughness. Therefore, further studies have been conducted with the aim of determining the texture of multilayers. The first studies carried out, which are mentioned in reference [13], have shown a diversification of the texture (the density of poles and the number of orientations) of multilayers.

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