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Full Length Article

Self-assembled metal nano-multilayered film prepared by co-sputtering method

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ABSTRACT

Nano-multilayered film is usually prepared by the arrangement deposition of different materials. In this paper, a self-assembled nano-multilayered film was deposited by simultaneous sputtering of Cu and W. The Cu/W nano-multilayered film was accumulated by W-rich layer and Cu-rich layer. Smooth interfaces with consecutive composition variation and semi-coherent even coherent relationship were identified, indicating that a spinodal-like structure with a modulation wavelength of about 20 nm formed during co-deposition process. The participation of diffusion barrier element, such as W, is believed the essential to obtain the nano-multilayered structure besides the technological parameters.

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1. Introduction

In recent years, nano-multilayered film has attracted extensive attention due to their practical and potential applications [1,2]. Until now, there are numerous studies about the mechanical, electronic, acoustic, thermal and many other properties of multilayers [3–5]. Multilayered film consists of alternating nanoscale layers containing the modulation period λ, modulation wavelength and content oscillation. Generally, the multilayered film has been prepared quite mature by deposition method with sequential deposition, adopting impulse power, e.g. impulse magnetron sputtering and pulsed laser deposition [6–8] or using shutters and/or movement of the substrate in front of separated sources [9]. Very few publications depicted the multilayers by keeping the substrate rotation during co-deposition process [10–14].

The substrate rotation and deposition time have been considered the main factors to form the multilayered structure during co-sputtering process [10–14]. For example, Plantin et al. [13]. obtained the Fe-W multilayered film by magnetron co-sputtering, which has the periodic oscillating composition at low substrate rotation rate and short deposition time. The periodic oscillation gradually disappeared as the rotation rate or deposition time increased. Müller et al. [10] and Chen et al. [12] also observed that the modulation period λ decreased as the substrate rotation rate increased. Chen et al. [12] further indicated that the number of the period was almost equal to the substrate revolutions. Other parameters, such as the substrate position on the sample holder and ion beam bombardment, can influence the amplitude of composition fluctuation and the formation of superlattices [11,15]. Among these investigations, the process parameters during co-sputtering deposition are responsible for forming the multilayered structure. However, Plantin et al. [13]. depicted that the formation mechanism was still unclear but not caused by heating or roughing. Why did just few specialization composition system form multilayered structure?

Excluding the process parameters, there may be other formation mechanism of the multilayered structure during co-sputtering deposition. Closely view the multilayered film, such as Cu-Ta [10], Ru-Ta [12], Fe-W [13], Ni-Pt [14], Ni-Au [15] and so on, obtained from the co-sputtering technique, we found an interesting character. That all these multilayered films contain a diffusion barrier element, such as W, Ta, Au, Pt, Ru. Once there are no diffusion barrier elements participation, no multilayered structure formed [16]. Similar multilayered structure, containing metal-rich (Me including Cu, Ag, Au) layer and a-C-rich layer, have been prepared by the co-deposition method [17]. Owing to the non-carbide forming elements (Cu, Ag, Au), it can be regarded as immiscible systems, in which the a-C can served as diffusion barrier element. How does the diffusion barrier elements induce the formation of multilayered

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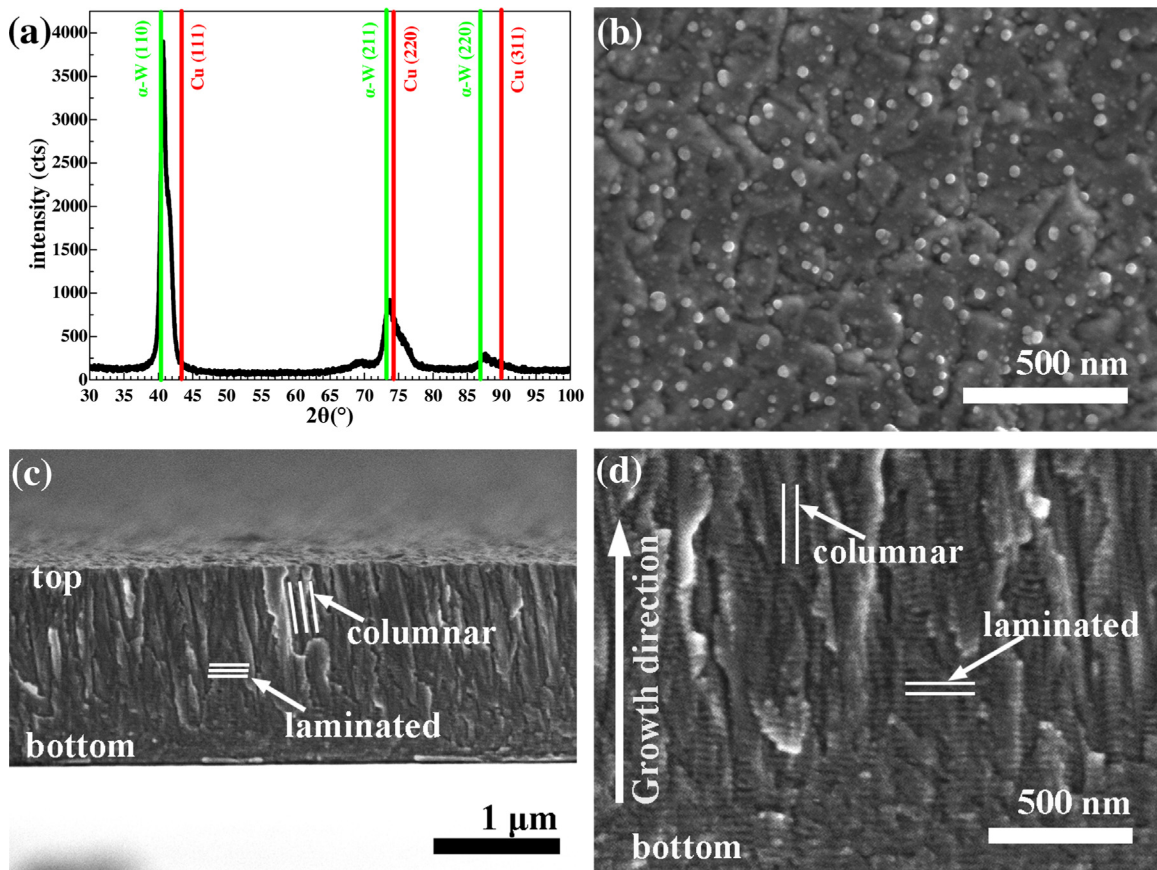

Fig. 1. (a) XRD pattern of the Cu-W film. SEM images of the sample (b) in planar and (c) in cross-section, (d) local high magnification.

structure during co-sputtering deposition? There is no idea as far as we know. So, in the present work, the W was selected as a diffusion barrier element, and Cu-W film was co-deposited by co-sputtering technique. The microstructures were analyzed by XRD, SEM and TEM. Within the films, multilayered structure was observed and the essential conditions of self-assembled nano-multilayered film were discussed.

2. Experimental

The Cu-W film was deposited on Si (100) single crystal by dual direct current magnetron co-sputtering system (MIS800), which is equipped with two independent and symmetrical magnetron targets. The sputter guns were 30° titled and focused on the substrate-holder. Metal targets of Cu (99.99%) and W (99.95%) disks with diameter of 50 mm were adopted as source materials for sputtering. The target to substrate distance was kept at 90 mm. The based pressure in the vacuum chamber was below 5×10^{-4} Pa, followed by the inlet of argon gas as a plasma source. The gas flow rate was 60 sccm causing the working pressure at 1.0 Pa. The Si substrate was cleaned by sputtering with 600 V argon ions for 15 min before deposition. The deposition time was 30 min with rotation speed of 2.5 rpm (rpm). The deposition rate and the composition of the thin films were controlled by the power of Cu and W targets. Annealing of the samples were performed at 150, 400, 600, 650, 700 °C for 1h. The samples were put into the sealed quartz glass tubes, which were vacuumized and filled with argon and titanium plates in order to eliminate oxygen. The heating rate to anneal temperature was 5 °C/min in all cases, and annealing was carried out in a thermal annealing furnace with flowing argon atmosphere. Nanoindentation tests on the films were performed using a TriboIndenter from Hysitron Inc. with a Berkovich indenter. The films were tested under maximum depth of indentation of 200 nm, which is less than one-seventh of the total thickness, and repeated six times on each sample.

The crystallographic structure was investigated by using a Bruker-AXS D8 Advance XRD (X-ray diffractometer). Analysis of cross sectional and surface morphologies were performed by Hitachi S-4800 FESEM (Field Emission Scanning Electron Microscope). The films cross-section for SEM were break off directly without mechanical polishing. Chemical composition analysis of the film was conducted by FEI Quanta 200 SEM with EDX (Energy-Dispersive X-ray spectroscope). Further nanostructure and crystallographic analysis were examined by FEI Titan G2 60-300 TEM (Transmission Electron Microscope) at a 300-kV accelerating voltage. The point resolution of TEM is 80 pm and the STEM resolution is better than 136 pm. The cross-section films for TEM observation were prepared by means of ion milling on a Gatan 691 PIPS (Precision Ion Polishing System). The voltage was controlled between 3–6 kV and the grazing incidence of Ar^+ ion beam was 1∼4° to the surface.

3. Results and discussions

3.1. *The analysis of the nano-multilayered structures and their interfaces*

Fig. 1a shows the XRD spectra of the Cu-W film. The diffraction peaks shift to higher angle show that the formation of W-based solid solution [18]. The broadened diffraction peaks and the weaker peaks on the shoulders indicate that continuous solid solution with different composition could be formed. It is noteworthy that the

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