Formation of Crystalline Copper Thin Films by a Sputtering-assisted Magnetic Field System at Room Temperature

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Abstract A sputtering-assisted magnetic field system was successfully developed for depositing crystalline Cu thin films at room temperature. This system employs a plasma source and an ion-beam gun with two magnetic field generators, which is covered with sputtering target and the ion-beam gun, simultaneously serving as sputtering plasma and a magnetic field generator. The formation of crystalline Cu thin films at room temperature was dominated by magnetic fields, which was revealed by preliminary experiments. This system can be employed for producing crystalline metal thin films at room temperature

Keywords: Copper thin film, Crystalline, Sputtering, Magnetic field

I. Introduction

Thin metal electrodes are essential components of numerous electronic devices [1,2]. Fabrication of thin metal electrodes is particularly important for flexible and wearable electronics to obtain superior electrical conductivity and to generate durable metal electrodes that undergo harsh bending or stretching conditions. The electrical conductivity and durability of metal electrodes could be enhanced by increasing their crystallinity [3-5]. Sputtering deposition is an inexpensive, efficient, and highly reproducible technique for fabricating thin metal electrodes, which has been widely adopted in several applications [6-8]. In general, high temperature during deposition is necessary to increase crystallinity [9]; however, the possibility of applying high-temperature fabrication conditions to flexible substrates is limited. Therefore, exploring suitable sputtering conditions for obtaining superior crystalline metal thin films at low temperature has is highly vital [10].

In this research, we fabricated Cu thin films that are widely employed as electrode materials in electronic devices using an ion-beam-assisted sputtering technique under a magnetic field. We studied the effect of magnetic fields on the crystallinity of the deposited Cu thin films. Cu thin film deposition by magnetron sputtering at low temperature enabled superior crystallinity and enhanced the electrical properties of the material.

II. Experiments

1. Fabrication of Mo and Cu thin film

All slide-glass and Polyethylene terephthalate(PET) substrate were initially cleaned by scrubbing with a soft cloth and a Triton X-100 solution, followed by ultrasonic cleaning in successive solutions of Triton X-100, water, and ethanol for at least 10 min each.

Scheme 1 shows the schematic drawing of a new sputtering system like the one used in our experiments. New sputtering system was comprised of dc magnetron gun, ion beam source, solenoid 1, and solenoid 2.

Metal molecules were sputtered from a circular polycrystalline metal target with the diameter of 2 inch. The base pressure of the deposition chamber was around \(10^{-6}\) torr, while the working process pressure was around \(4\times10^{-3}\) torr. Pure argon gas (purity: 99.999%) was used as the inert gas in the chamber. Argon plasma was applied at a dc power of 150 W. The distance between target gun and substrate was fixed 16 cm. Also, the distance and incident angle (from the standard of substrate horizontality) between ion beam source and substrate was fixed 19 cm and 60 degrees, respectively. We deposited a metal film layer on a substrate by using the dc magnetron sputtering technique at room temperatures. In our experiment we adjusted the thickness of the deposited films to 100 nm. Solenoid 1 dc power was controlled from 0 to 31.5 W (21 V, 1.5 A). Sputtering ion metal atom was flown to keeping ion state on substrate surfaces due to magnetic field of Solenoid 1.

The substrate surfaces were bombarded by an argon ion beam. A cold-hollow-cathode (CHC)-type ion source was
used to yield the ion beam. In order to collimate the ion beam, we used two perforated grids as electro-focusing lenses. The CHC-type ion source is composed of a separately cooled chamber supplied with a magnetic system and is connected to a discharge chamber through an orifice. Argon gas is fed into the ion source through the CHC only. The discharge ignition in the cathode takes place at a nominal discharge voltage and gas flow rate. Metal film surfaces was exposed to an ion beam as a function of exposure time (ion beam energy, 30 eV; incident angle, 60 deg; ion beam flux density, \(1.5 \times 10^{13} \text{ Ar}^+ / \text{s} \cdot \text{cm}^2\)). Solenoid 2 dc power was controlled from 0 to 15 W (15 V, 1 A). Solenoid 2 was played the role of flying in linearity argon gas to substrate surfaces due to magnetic field of Solenoid 2.

2. Characterization of Cu thin film

XRD measurements of the metal film was performed using an X-ray diffractometer (X'Pert Pro, Philips, Netherlands) equipped with a monochromatic Cu Kα radiation (\(\lambda=1.054056 \text{ Å}\)) operated at 40 kV and 30 mA. The diffraction pattern was measured at room temperature in normal θ-2θ scanning mode over angles ranging from 10° to 90° with a step of 0.05° and a continuetime of 0.2 s/step. We also characterized the film's surface morphology by using AFM with the tapping mode (Digital Instruments, Multimode AFM Nanoscope IIIa). An ultra-lever cantilever with a spring constant of 26 N/m and a resonance frequency of 268 kHz was used for scanning. Measurements were performed using the four-point probe test for the determination of resistivity of thin films. Rectangular sections (2 cm × 3 cm) were cut from the substrate. The surfaces of the sections were cleaned thoroughly before the resistivity measurements were made. A Keithley 2400 sourcemeter was used to source current (1 mA) through the sample; the voltage drop across the precursor was measured by a Keithley 2182 nanovoltmeter. The specific resistance was determined by taking into account thin film shape and thickness. The electrical measurements took place in a defined atmosphere: vacuum (10\(^{-1}\) mbar) and N (5 N).

III. Results and Discussion

Figure 1 illustrates a detailed cross-sectional structure schematic of our objective sputtering apparatus equipped with an ion-beam gun (denoted as \(I_g\)). Two solenoid conductors were employed near the metal target [denoted as \(S(1)\)] and the ion beam gun [denoted as \(S(2)\)]. In this study, we focused on the effect of the magnetic field, generated by the solenoid conductors \(S(1)\) and \(S(2)\), on the crystallinity of the sputtered Cu thin film on the glass substrate.

X-ray diffraction (XRD) patterns of the as-deposited Cu thin films, obtained at room temperature under the indicated sputtering conditions, are displayed in Fig. 2. As can be seen, under deposition conditions in the absence of a magnetic field, the as-deposited Cu thin films did not exhibit any diffraction peak, indicating a lack of crystallinity. After Cu deposition under simple ion-beam irradiation on the sample substrate, relatively weak and broad diffraction peaks were observed at 20 values of 29.2° and 60.6°, which correspond to the (002) and (200) planes of CuO due to oxidation of the Cu surface.

Remarkably, when the Cu thin films were deposited by sputtering using a magnetic field from \(S(1)\) at room temperature, distinct XRD diffraction peaks could be observed at 20 values of 43.5°, 50.6°, and 74.3°, which correspond to the (111), (200), and (220) planes, as shown in Fig. 1.
in Fig. 2. The appearance of these XRD patterns confirmed the excellent crystallinity of the deposited thin films at a working electric field from S(1). Moreover, when the deposition was performed under S(1) with simultaneous ion-beam irradiation from S(2), the intensities of the three diffraction peaks increased significantly compared with the results obtained from other conditions. This result indicates that the electric fields from S(1) and S(2) enhance the crystallinity of the deposited Cu thin films at room temperature. No crystalline Cu thin films were formed in the absence of S(1) and S(2) at room temperature, indicating that the presence of a magnetic field is mandatory for crystallization at room temperature.

Atomic force microscopy (AFM) was performed to analyze the root-mean-square (RMS) roughness of the films. Figure 2 shows the AFM images of the as-deposited Cu thin films on a glass substrate obtained under various deposition conditions (as indicated). The AFM images of the deposited Cu thin films as a function of the magnetic field show a compact granular structure.

For the various Cu thin films deposition conditions such as those without ion-beam irradiation and magnetic field, with ion-beam irradiation only, with the magnetic field S(1) alone, and with magnetic fields S(1) and S(2) under ion beam irradiation, the corresponding RMS roughness values were 12, 13, 25, and 32 nm, respectively. The RMS roughness values increased with an improvement in crystallinity.

To explore the effect of the magnetic field on the quality of the thin films, we studied the electrical conductivity of the deposited Cu thin films at different strengths of the magnetic field S(1) at room temperature. In the absence of a magnetic field from S(1), the resistivity ($\rho$) of the Cu thin films was $\rho = 35 \mu \Omega \cdot \text{cm}$, which gradually decreased to $4.6 \mu \Omega \cdot \text{cm}$ as the magnetic-field strength S(1) increased, as shown in Fig. 3. This trend may be attributed to the enhancement of crystallinity, as shown by the XRD peaks.

**IV. Summary**

Crystalline Cu thin films could be achieved at room temperature by employing a sputtering system equipped with magnetic field generators S(1) and S(2). We could subsequently prove that both magnetic fields S(1) and S(2) significantly affected the film-crystallinity control, so a fine control might be possible by using the current correction from the field calculations. This technique is expected to improve crystalline metal-electrode deposition at room temperature, enabling a series of metal-deposition processes for the fabrication of metal electrodes on various plastic substrates.

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