2Pa1-1

Proceedings of Symposium on Ultrasonic Electronics, Vol. 32 (2011) pp. 57-58
8-10 November, 2011

In-situ Observation of Film Growth during Sputtering by Ultrasound Spectroscopy.

超音波スペクトロスコピーによるスパッタリング中の薄膜成長その場観察

Junki Okamoto[1‡] , Nobutomo Nakamura[1], Hirotsugu Ogi[1] , Masahiko Hirao[1] and Masayoshi Nishiyama[2] ([1]Graduate School of Engineering Science, Osaka Univ.; [2]Renovation Center of Instruments for Science Education and Technology, Osaka Univ.)

岡本 純基 [1‡], 中村 暢伴 [1], 荻 博次 [1], 平尾 雅彦 [1], 西山 雅祥 [2] ([1] 大阪大学大学院基礎工学研究科, [2] 大阪大学科学教育機器リノベーションセンター)

1. Introduction

Elastic stiffness of polycrystalline thin films tends to be smaller than that of the corresponding bulk materials. This is thought to be caused mainly by defects at grain boundaries that were formed during the deposition[1]. In polycrystalline Cu thin films, elastic stiffness recoveries to the bulk value by annealing[2]. The recovery is consistently explained by the reduction of defects, and this result supports the view that defects soften polycrystalline thin films.

Understanding the film-growth process is an important task to reduce the defects and improve the elastic property. X-ray diffraction[3] and curvature measurement[4] have been used for studying the structural evolution during and after the deposition. However, relationship between the structural evolution and the configuration of defects has not been discussed, because those techniques were insensitive to the defects existing at grain boundaries.

For solving this difficulty, we have developed an acoustic method[5]. It succeeded to detect the volume-fraction change of defects after the deposition, and recovery of elastic stiffness by 10 % was observed[6]. However, structural change in a very early stage is still unknown, because the method was insensitive to ultrathin films (less than ten nanometer thick). In the early stage, drastic structural evaluation has been observed[7], and it should contribute the final configuration of the defects. In this study, a method developed for observing the structural change in the early stage is shown. Then, experimental results on Pt films are discussed.

2. Experimental Procedure

The method is based on the resonant-ultrasound spectroscopy (RUS)[8,9]. Resonance frequencies of a substrate on which a thin film is deposited depend on the dimension, mass density, and elastic stiffness of the substrate and film[10]. During deposition at a constant deposition rate, the resonance frequencies should change monotonically. However, if drastic structural change happens and elastic stiffness is consequently changed, the resonance frequencies change anomalously. Therefore, by monitoring the evolution of the resonance frequencies during deposition, structural change is detected. In previous study, we used a silicon substrate (100 µm thick) and a tripod-piezoelectric transducer[6]. Using smaller substrate enables us to detect smaller change in the structure. However, the tripod-piezoelectric transducer could not detect the structural evolution in ultrathin films. In this study, we solve this by using a quartz crystal (1.7 mm × 2.5 mm × 9 µm) as a substrate and applying oscillating electric field to it. We set them in RF magnetron sputtering chamber (**Fig.1**), and

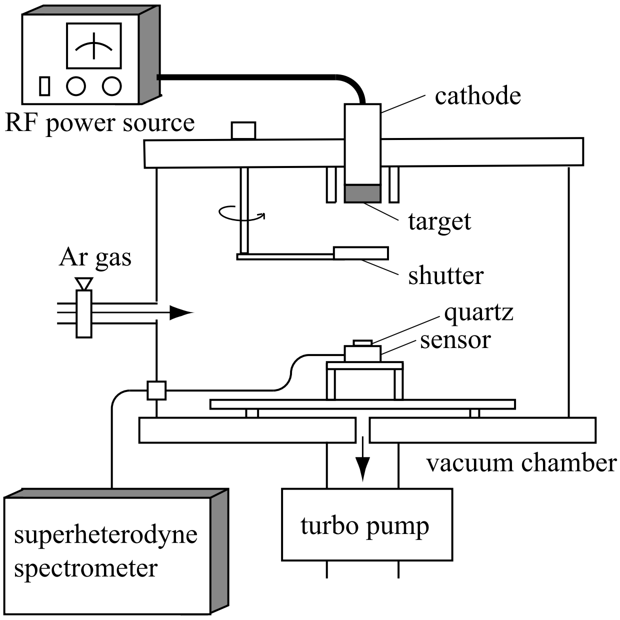

Fig. 1 Schematic of RF Magnetron sputtering chamber and measurement system.

Email: junki.okamoto@mbox.me.es.osaka-u.ac.jp

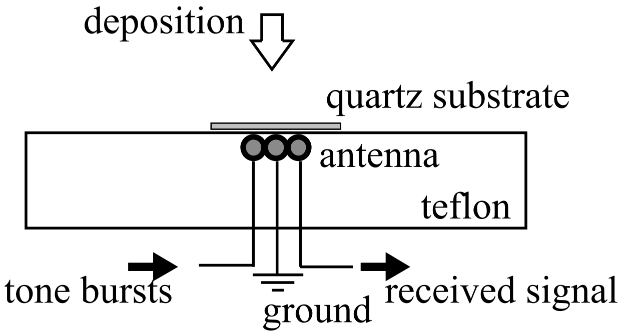

Fig. 2 Schematic image of the sensor. Noncontact antennas transmit and detect the vibration.

measured a resonance frequency during and after deposition.

Fig.2 shows the schematic image of the acoustic sensor. Quartz substrate is vibrated by electric field which antenna excites. The antenna consists of three straight copper wires, two for generation and detection of the mechanical free vibrations and the other for the grounding wire. The copper wires are embedded in the top face of the sensor cell made of Teflon. By applying the tone bursts around resonance frequency to the generation wire, the electric field occurs in-plane parallel to the surface. This filed generates plane waves propagating in the thickness direction in a substrate through the converse piezoelectric effect. After the excitation, the detection wire detects a quartz vibration by the piezoelectric effect. The received signal is send to the superheterodyne spectroscopy, and the Gaussian function fitting provides the resonance frequency. **Fig. 3** shows a typical resonance spectrum of the quartz substrate by this method. Because there's no need to directly attach substrate on antenna, we can measure the frequency without affecting deposition. Piezoelectric effect arises only in a resonant condition, so we can measure resonant frequency. Background pressure in the sputtering chamber was less 2.0×10^{-4} Pa. Ar pressure was 0.4 Pa. Pt was deposited at room temperature and deposition power was 10 W. Sputtering rate was 8.2 nm/h.

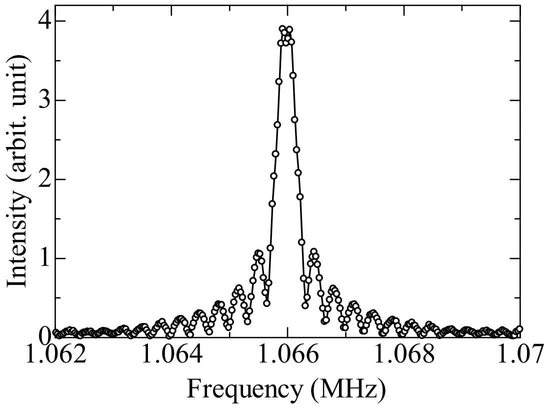

Fig. 3 Measured resonance spectrum of the quartz substrate.

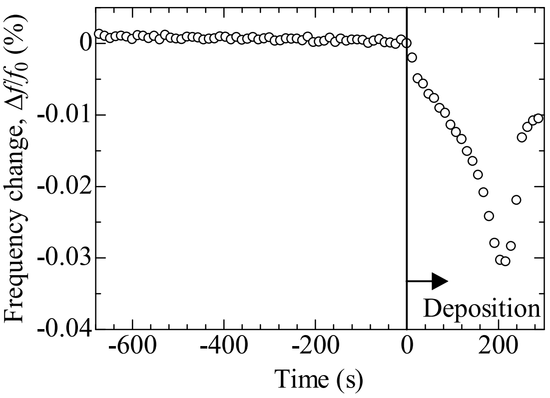

Fig. 4 Change in the resonance frequency of a quartz substrate before and during sputtering of Pt. At 0 s, shutter was opened and deposition was started

3. Results and Discussion

Fig. 4 shows the change ratio of the resonance frequency by the deposition of Pt. The resonance frequency of the quartz substrate before the deposition was 3.3707 MHz. The frequency decreased after starting the deposition and started to increase at 200 s. At that time, film thickness was 0.46 nm. Decrease of resonance frequency during the deposition was an expected result because of increase of mass by deposition[10]. But increase of the resonance frequency cannot be explained by the change in the mass.

We consider that stiffening of the Pt film, increase of the stiffness, is the possible reason. In metallic thin films, morphology change that isolated islands coalesce to form continuous film occurs. Then, the structural change could cause significant increment of the stiffness. We consider that observed frequency change reflects the coalescing.

References

1. N. Nakamura, H. Ogi, and M. Hirao: Acta Mater. **52** (2004) 765.
2. N. Nakamura, H. Ogi, T. Syagawa and M. Hirao: Appl. Phys. Lett. **92** (2008) 14901.
3. J. W. Pattern, E. D. McClanahan and J. W. Johnston: J. Appl. Phys. **42** (1971) 4371.
4. J. A. Floro, S. J. Hearne, J. A. Hunter and P. Kotula: J. Appl. Phys. **89** (2001) 4886.
5. N. Nakamura, H. Ogi, T. Nakashima, M. Hirao, and M. Nishiyama: Jpn. J. Appl. Phys. **46** (2007) 4450.
6. N. Nakamura, T. Nakashima, H. Ogi, M. Hirao, and M. Nishiyama: J. Appl. Phys. **107** (2010) 103541.
7. R. N. Esfahani, G. J. Maclay, G. W. Zajac: Thin Solid Films **219** (1992) 257.
8. I. Ohno: J. Phys. Earth **24** (1976) 355.
9. A. Migliori, J. L. Sarrao, W. M. Visscher, T. M. Bell, M. Lei, Z. Fisk, and R. G. Leisure: Physica B **183** (1993) 1.
10. P. Heyliger: J. Acoust. Soc. Am. **107** (2000) 1235.