2P1-4
Proceedings of Symposium on Ultrasonic Electronics, Vol. 34 (2013) pp. 195-196
20-22 November, 2013

Monitoring of Film Growth on Quartz by Acoustic Resonance Method
超音波共振法による水晶上での薄膜成長観察

Naoto Yoshimura[‡], Nobutomo Nakamura[*], Hirotsugu Ogi, and Masahiko Hirao
(Graduate School of Engineering Science, Osaka Univ.)
吉村尚人[‡]，中村暢伴[*]，荻博次，平尾雅彦（大阪大学大学院基礎工学研究科）

1. Introduction

Nowadays there are numerous applications using thin films and it has become an essential material for modern engineering. With the technology progress, the demand for high quality thin film is getting larger. For example, the improvement of uniformity and decrease of thickness of the thin film leads to miniaturization of the device such as semiconductor. Understanding the dynamics of the film growth and accurate control of the film growth are then important research topics. Polycrystalline metallic thin film, generally used for such devices, often shows different elastic property from the corresponding bulk materials [1, 2]. This difference comes from the defective structures inside the film introduced during the deposition. The defects are to change during and after the deposition, and the volume fraction and shape vary with the deposition condition. Therefore, for understanding the formation of the defective structure, monitoring the structural evolutions in real-time is required.

Structural evolutions during the deposition have been investigated by various measurements such as scanning tunneling microscopy [3] and curvature measurements [4]. Scanning tunneling microscopy can observe the structural evolution directly, but cannot monitor rapid structural evolution. Curvature measurement can measure stress evolution throughout the deposition by applying the Stoney's equation [5] to the measured curvature of the substrate. However, residual stress is affected by elastic stiffness in addition to the structural evolution, and it makes analyzing the film structure from the stress difficult. For these reasons, we propose the Antenna Transducer Acoustic Resonance (ATAR) method for monitoring the structural evolution. ATAR investigates the properties of deposited film from the resonance vibration of piezoelectric substrate. Resonance frequencies of piezoelectric materials depend on dimensions, density, elastic stiffness, piezoelectric constants, and dielectric constants. When a thin film is deposited on the substrate, resonance frequencies change depending on the thickness, density, and elastic stiffness of the film. Furthermore, deposition of a metallic film affects the electrical boundary condition of the piezoelectric substrate, and evolution of the electric conductivity of the film will be obtained from the vibrational properties. Measurement of the internal friction also gives dynamic information of the film growth. For these reasons, the ATAR possesses potential to observe evolution of several properties that cannot be observed by the other methods.

2. Method

The resonance frequency, vibrational amplitude, and internal friction are determined by measuring the resonance spectra with the antenna. **Fig. 1** shows the schematic drawing of the antenna. Oscillating electric field generated by tone burst excites acoustic vibration in the piezoelectric substrate by the inverse piezoelectric effect. Electric field excited by the acoustic vibration of the substrate is detected by the antenna, and by sweeping the frequency of the tone burst, a resonance spectrum is obtained. Substrate is just placed on the antenna, and no external force except gravity acts on the substrate, achieving ideal free vibration.

The antenna and quartz substrate were placed in a vacuum chamber. Film was deposited by RF magnetron sputtering, and deposition rate was determined from the duration time of the deposition and film thickness determined by x-ray reflectivity technique. Base pressure was less than 3×10^{-4} Pa. Ag thin film was deposited on *z*-cut quartz substrate measuring 2.5 mm, 1.7 mm, and 200 μm in the *x*, *y*, and *z* direction, respectively.

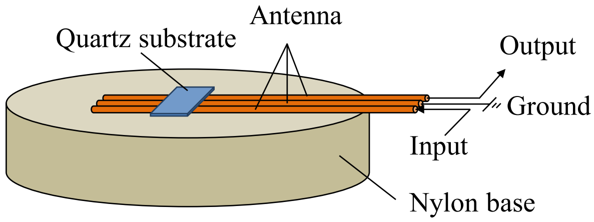

Fig. 1 Schematic drawing of the antenna.

[*]nobutomo@me.es.osaka-u.ac.jp

3. Results and discussion

In the monitoring, one of the resonance frequencies is focused. **Fig. 2** shows the typical result of change ratio of the resonance frequency and amplitude during the Ag deposition on 200 μm quartz substrate. The resonance frequency before the deposition was 1.079 MHz. The result shows sharp decrease in both resonance frequency and amplitude around 800 s. It is considered that these decreases originate from the disappearance of piezoelectricity. Growth of Ag film on quartz is classified in the Volmer-Weber model (**Fig. 3**), in which the deposited atoms form three-dimensional island structure in the initial stage of growth. When it is isolated island, quartz is still able to polarize and the antenna can excite vibration. However, when the structure of the deposited film changes from isolated island to continuous film, the surface of the substrate became equipotential and it is not able to polarize anymore (see Fig. 3). This indicates that antenna cannot oscillate the substrate, and amplitude decreases drastically at that moment. In addition, the lack of the polarization leads to the lack of piezoelectricity. Apparent stiffness then decreases, and the resonance frequency decreases at the same time. We also measured the internal friction of the substrate during the deposition, and observed that the internal friction takes the maximum value when the resonance frequency and amplitude decrease sharply. Diffusion of the deposited atoms is one of the possible reasons for this increment, and diffusion of the deposited atoms could be activated when the deposited film changes from isolated island to continuous film.

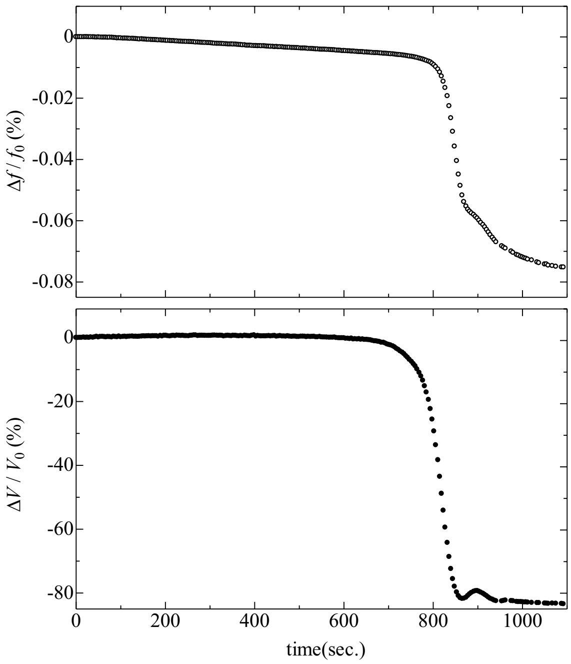

Fig. 2 Change ratio of frequency (top) and amplitude (bottom) during the deposition of Ag on the 200 μm quartz substrate. The resonance frequency before the deposition was 1.079 MHz.

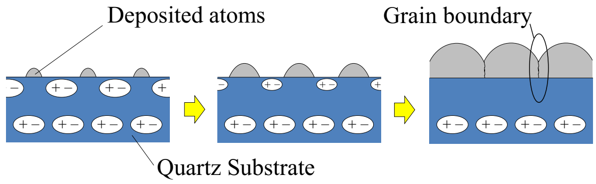

Fig. 3 Polarization near the surface of a quartz substrate during deposition in the Volmer-Weber model.

4. Conclusions

We succeeded in monitoring the film growth on quartz substrate by ATAR method. By this method the moment when the deposited film changes from isolated island to continuous film was observed by the sharp decrease of the resonance frequency and amplitude, and the increase of internal friction. This measurement is performed only by putting the specimen on the antenna. Though there is a restriction that it can only be applied on piezoelectric substrate, it can be a convenient method for film growth monitoring.

References

1. H. Ogi, N. Nakamura, H. Tanei, M. Hirao, R. Ikeda, and M. Takemoto: Appl. Phys. Lett. **86** (2005) 231904.
2. H. Ogi, M. Fujii, N. Nakamura, T. Shagawa, and M. Hirao: Appl. Phys. Lett. **90** (2007) 191906.
3. M. Poensgen, J. F. Wolf, J. Frohn, M. Giesen, and H. Ibach: Surf. Sci. **274** (1992) 430.
4. D. Flötotto, Z. M. Wang, L. P. H. Jeurgens, E. Bischoff, and E. J. Mittemeijer: J. Appl. Phys. **112** (2012) 043503.
5. G. G. Stoney: Proc. R. Soc. Lond. A **82** (1909).