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Preparation of new nitride/oxide nanocomposite coatings by a newly developed differential pumping cosputtering system

新開発の差動排気型２元同時成膜装置による酸化物／窒化物系ナノコンポジット膜の作製

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A new sputtering method, the differential pumping co-sputtering system (DPCS), was developed to controllably fabricate novel, multi-functional nanocomposite films with compositions unconstrained by thermodynamic restrictions. This system features a multi-chamber design with a differential pumping system. Dividing atmospheres to two chambers with narrow clearance (1–2 mm) greatly reduced the cross-contamination among chambers, and each material could be co-deposited by substrate rotation. OES measurement in the glow discharge plasma did not detect substantial oxygen contamination from the other chamber. Using this system, nanocomposite films such as AlN/SiO_x, AlN/Al_xO_y consisting of B4-type AlN and amorphous SiO_x or Al_xO_y were obtained.

1. Introduction

In order to fabricate nanocomposite films, multiple targets sputtering suffer from a limitation of the thermodynamic restriction on compound formation[1, 2]; provided that a metal “A” is more easily oxidized than another metal “B”, it is very difficult to fabricate nanocomposite films of A/B-O, or A-N/B-O (A, B: Metal; O: Oxygen, N: Nitrogen) that consist of metal A and an oxide B or nitride A and oxide B. To overcome such a problem and to fabricate novel, multi-functional nanocomposite films with compositions unconstrained by thermodynamic restrictions, we have developed a new sputtering system called the differential pumping co-sputtering system (DPCS).

2. System Design and Experimental

Figure 1 shows a schematic diagram of DPCS for a two-chamber design. The most important consideration in the design of our DPCS for nanocomposite films is being able to achieve two contradictory operations simultaneously. Dividing atmospheres reduced the cross-contamination among chambers, and each material could be co-deposited using rapid substrate rotation. Minimizing the clearance between the substrate holder and the chamber walls provides reduced cross-contamination between the chambers. However, for rapid rotation of the substrate holder, a margin of clearance was required in consideration of deformation of these parts. Consequently, we have designed and fabricated a clearance of ~2 mm between the two chambers in consideration of thermal expansion and deformation of the chamber by atmospheric pressure.

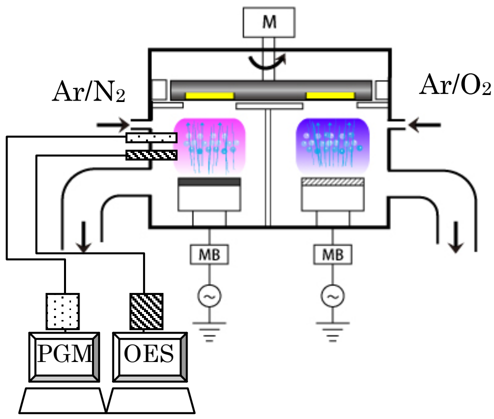

Fig.1 Schematic diagram of the differential pumping co-sputtering system (DPCS) consisting of two chambers. [3]

To examine the cross-contamination between the right and the left chambers, the change of oxygen partial pressure in the left chamber was measured with the introduction of different gases to each chamber using a PGM. And, we analyzed the glow discharge plasma in the left chamber by OES under different discharge conditions in the right chamber. Furthermore, we tried to fabricate a new nanocomposite film consisting of AlN and SiO_x using DPCS. The crystal structure of the films was identified by an x-ray diffractometer using Cu-*K*α radiation with θ-2θ or grazing angle mode (GAXRD). The microstructure of the films was investigated by TEM.

3. Results and Discussion

The physical framework necessary to adequately describe the gas flow depends on the Knudsen Number, *Kn*, which is defined as the ratio of the mean free path (MFP) between gas atom/molecule collisions (λ) and a characteristic dimension of the gas flow system under consideration (*D*). The conditions for molecular and viscous flow suggest that the former occurs when $Kn > 1$ and the latter, when $Kn < 0.1$.

Molecular flow: $$Kn = \lambda / D > 1 \quad (1)$$
Viscous flow: $$Kn = \lambda / D < 0.1 \quad (2)$$

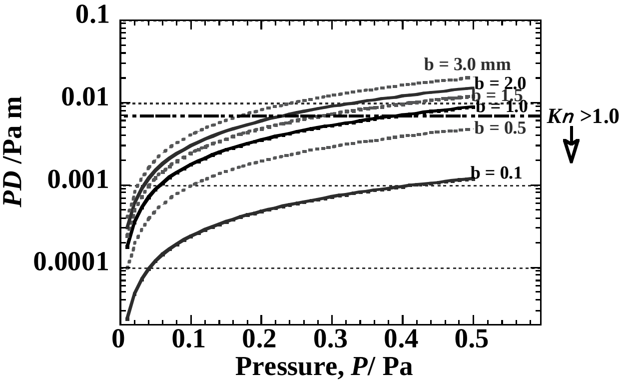

Fig. 2 The PD value vs. sputtering gas pressure as a function of clearance for the DPCS.[3)]

From the equation of Knudsen number, *Kn*, the PD values can be defined.[4)] PD values were calculated from Knudsen number and conductance of clearances[5)]. Figure 2 shows the variation of the PD value with sputtering gas pressure as the clearance (b) is varied from 0.1 to 3.0 mm. The PD values decreased with the decrease of sputtering gas pressure for each clearance; in particular, the values decreased drastically under gas pressures lower than 0.1 Pa. However, normal sputtering gas pressure is higher than 0.1 Pa. To satisfy the criterion of the molecular flow condition at 0.3 Pa, the clearance must be less than 1.5 mm. Although we have not measured the actual clearance during sputtering, the clearance is expected to be reduced to 1.0–1.5 mm by atmospheric pressure and thermal expansion, and then, the conductance of the PD value at 0.3 Pa seems to be within the molecular flow regime. The PGM results revealed that the cross- contamination of oxygen from the other chamber was suppressed to 10±3 % of the original. The OES measurement in the glow discharge plasma did not detect substantial oxygen contamination from the other chamber.

A new nanocomposite film consisting of nitride (AlN or CrAlN) and oxide (Al_xO_y or SiO_x) were fabricated using the DPCS. In the left chamber, nitride deposited under a mixture of Ar/N_2 atmosphere at 0.3Pa and, in the right chamber, oxide was deposited under an Ar gas of 0.2Pa, respectively. The targeted mixing ratio were 0~50 vol% of oxide as determined from the sputtering rate of each film. XRD patterns of the obtained films revealed the presence of only one phase that can be assigned to the hexagonal B4 structure (AlN) or cubic B1 structure (CrAlN). No other peaks corresponding to other compounds or unknown phases were detected.

4. Conclusion

We have developed a unique sputtering system named as the differential pumping co-sputtering system (DPCS) to reduce the cross-contamination between chambers. We roughly calculated and examined the Knudsen number of the clearance between two chambers using a model orifice. The calculation showed that the cross-contamination was suppressed even under different gas pressures between the two chambers with the aid of a practical molecular flow. By PGM/OES experiments, we showed that while the cross-contamination was not null, it was suppressed to a considerable extent. We have also demonstrated that this system could be used to prepare new nanocomposite films consisting of nitride (AlN or CrAlN) and oxide (AlxOy or SiO_x) by the co-sputtering of metal (Al or CrAl alloy) and oxide (Al_2O_3 or SiO_2) targets under different atmospheres in each chamber of this system.

Acknowledgments

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