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Structural Control of Sputter Deposited Films beyond the Thornton Diagram Using Impurities

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Structures of sputter-deposited films have been mostly determined in a way shown in the Thornton diagram, in which there are two key parameters: substrate temperature and sputtering pressure. Here we add another parameter, impurities, that adsorb on, migrate on, desorb from the surfaces, and eventually modify the film structures.

The first example is the growth of amorphous In_2O_3:Sn (a-ITO) films. a-ITO films have attracted attention due to the advantages such as surface smoothness, low internal stress, and high etching rate. Conventional a-ITO films fabricated below crystallization temperature of In_2O_3 (150°C), that is Zone-1 in the Thornton diagram, however, possess high resistivity over 5×10^{-4} Ω·cm and poor thermal stability, limiting the use of a-ITO films in practical devices. Interestingly, we have observed that sputtering with nitrogen enables us to grow a-ITO films at extremely high temperature of 600°C (Zone-T in Thornton diagram) (Fig.1) and that the films have high mobility and high carrier density and thus low electrical resistivity of 2.6×10^{-4} Ω·cm (Fig.2), which is comparable to that of polycrystalline ITO films. This is because the high-temperature deposition leads both to increased film density and to enhanced short- and mid-range ordering in the amorphous structures. We found from x-ray fluorescence measurements that despite the large flow rate ratio of N2, where N atom density in the deposition atmosphere is one order of magnitude higher than O atom density due to the high dissociation degree of N_2 [2], the nitrogen contents in the films are only a few at.%. These results suggest that the amorphization is caused not only by the nitrogen atoms incorporated in the films, but by the adsorbed nitrogen atoms that inhibiting the crystal nucleation, which are eventually desorbed from the surfaces. Owing to the amorphous structure, the a-ITO films fabricated with nitrogen at 600°C have smooth surfaces with the root mean square (RMS) roughness of 0.6 nm, which is one third of that of films fabricated without nitrogen.

The second example is the growth of single crystalline ZnO films on 18%-lattice mismatched sapphire substrates, where nitrogen atoms play important roles to modify the structures again [2]. Sputtering with nitrogen brings about nm-sized grains even at high temperature over 700°C, which is less than one-tenth of the grain size of the films fabricated without nitrogen. The resultant films consisting of such small grains serve as quite-effective buffer layers because of the high degree of in-plane orientation and strain-relaxed smooth surfaces. In fact, we observed heteroepitaxial growth of ZnO films on sapphire substrates, where the films has single-crystalline structures with atomically-flat surfaces. This is a groundbreaking achievement that offer many more combinations of thin film materials and substrates for single-crystal film growth.

We believe that sputtering deposition with "impurities" offer new opportunities for designing materials with unprecedented structures.

References

[1] T. Takasaki, et. al., Proc. 9th ICRP, 60, GT1.150 (2015).
[2] N. Itagaki, et al., Sci. Rep., 10, 4669 (2020).

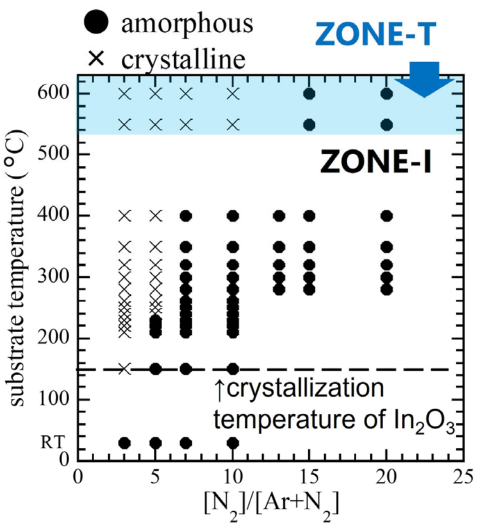

Fig. 1. Structural phase mapping of ITO films in the temperature-N_2 flow rate ratio plane.

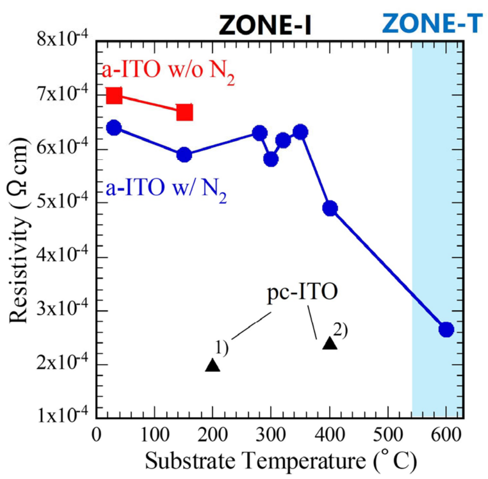

Fig. 2. Electrical resistivity of ITO films plotted against the substrate temperature.