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Non-Equilibrium and Extreme State -High-mobility amorphous In_2O_3:Sn films prepared via nitrogen-mediated amorphization-

非平衡極限　−窒素添加法による高移動度・非晶質In2O3：Sn 膜の作製−

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We have developed a fabrication method of amorphous In_2O_3:S_n (ITO) films, “nitrogen mediated amorphization (NMA)”, where magnetron sputtering is employed for film deposition. By utilizing this method, amorphous ITO films with smooth surface, where the RMS roughness of 0.28-0.47 nm, have been fabricated at various substrate temperatures in the range 150-400°C. The most remarkable feature of the NMA-ITO films is the high carrier mobility. In spite of the amorphous nature, the mobility is high of around 50 cm^2/Vs, being independent of the substrate temperature, which is about 2-8 times higher than that of conventional poly-crystalline ITO films.

1. Introduction

Tin-doped indium oxide (ITO) films having high transparency in the visible region along with the low electrical resistivity are widely used in flat panel displays, touch panels, solar cells, and other optoelectronic devices. Recently, amorphous ITO (a-ITO) films, which are generally obtained by lowering the deposition temperature, have attracted attention due to their superior properties such as smooth surface, high etching rate, and low internal stress. Such amorphous phases, however, is unstable and it can easily crystallize even at low temperature (~150°C), limiting the use of a-ITO films in practical devices. Therefore, it is of great importance to develop fabrication methods of ITO films with stabilized amorphous structure.

Recently, we have developed a new method of crystal growth based on magnetron sputtering, “nitrogen-mediated crystallization”, where crystal nucleation and the growth are controlled by nitrogen introduced as impurities [1-6]. Since the crystallization of ITO films is also expected to be inhibited by the existence of impurities in the films, addition of certain amount of nitrogen can disorder the crystal structures and thus bring ITO films with amorphous structures.

Here, we demonstrate “nitrogen-mediated amorphization (NMA)” of ITO films, where amorphous structures are obtained by introducing N_2 gas into the sputtering atmosphere instead of lowering the deposition temperature. The effects of NMA method on the morphology, crystallinity, and electrical properties of ITO films are discussed with comparison to poly-crystal ITO films fabricated by a conventional method.

2. Experimental

ITO films were deposited on quartz glass substrates by RF magnetron sputtering. The target-substrate vertical distance was 65 mm. The supplied RF power was 100 W and the substrate temperature was 150-400°C. Ar-N_2 mixed gas with N_2/Ar flow rate ratio of 0-20% was used and the total gas pressure was 0.9 Pa.

The thickness of the ITO films was 50 nm, confirmed by X-Ray-Reflectometry (Bruker D8 Discover). The electrical properties and the crystal structures of the films were evaluated by hall measurements and glancing angle X-ray diffraction (GXRD) analyses, where the incident angle α is fixed at a small angle (α=1.0°) so that X-rays are focused on the topmost surface of the sample. The surface morphology was studied by atomic force microscopy (AFM).

3. Results and Discussion

Figure 1 shows the GXRD patterns of as-deposited ITO films fabricated at 400°C at various N_2/Ar flow rate ratios. The diffracted peaks at 2 theta of 30.2° and 35.2°, which are correspond

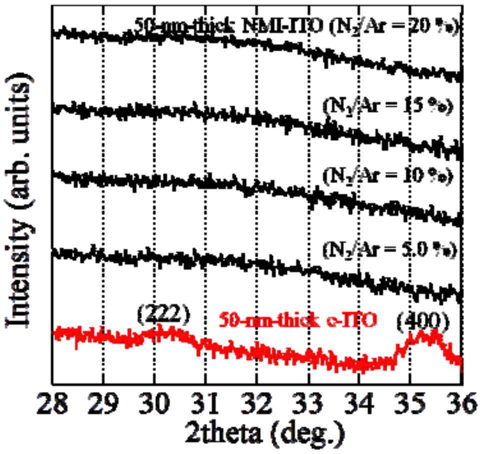

Fig. 1. GXRD spectra of ITO films prepared at various N_2/Ar flow rate ratios. c-ITO denotes the films prepared by conventional sputtering without nitrogen.

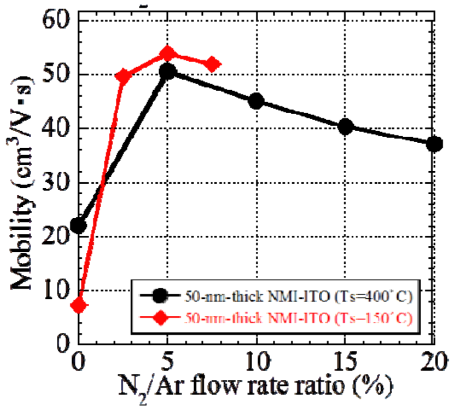

Fig. 2. Electron Hall mobility of ITO films measured at room temperature as a function of N_2/Ar flow rate ratios. The films were fabricated at 150°C and 400°C.

to the (222) and (400) planes of In_2O_3 bixbyite structure, were observed for conventional ITO films (c-ITO) prepared without nitrogen. On the other hand, no peaks are observed for ITO films prepared by NMA method, indicating that all of the NMA-ITO films are amorphous. AFM analysis revealed that the surface roughness for the NMA-ITO films is very small due to the amorphous nature. The RMS roughness of NMA-ITO films is in the range 0.32-0.45 nm for various deposition condition, whereas the RMS roughness of c-ITO films is around 1.3 nm. Furthermore, we found that the NMA-ITO films maintain the amorphous feature even after they were annealed at 400°C for 3 hours both in air and in N_2 atmosphere. From these results, we conclude that the existence of nitrogen in ITO films stabilizes the amorphous structure by inhibiting the crystallization of In_2O_3 into the bixbyite structure of a thermodynamically stable crystalline phase.
Figure 2 shows the hall mobility of the ITO films deposited at 150 and 400°C as a function of N_2/Ar flow rate ratio during deposition. The electron Hall mobility of the ITO films is significantly increased by N_2 introduction into the sputtering atmosphere. The highest mobility of the NMA-ITO films was obtained at N_2/Ar flow rate ratio of 5%, where the Hall mobility was around 50 cm^2/Vs, being independent of the deposition temperature. On the other hand, the mobility of c-ITO films was low of 7 and 22 cm^2/Vs at the deposition temperature of 150 and 400°C, respectively. As a results, a-ITO films with low resistivity of 3.4×10^{-4} was obtained in spite of the amorphous nature. These results allow us to conclude that NMA method is a powerful method of fabricating high-quality a-ITO films.

4. Conclusions

We report here a novel fabrication method of a-ITO, "nitrogen-mediated amorphization (NMA)". By utilizing this method, a-ITO films with stabilized amorphous structure have been successfully fabricated at various substrate temperatures in the range 150-400°C. The resultant a-ITO films have smooth surface, where the RMS roughness is in the range 0.32-0.45 nm. The electron Hall mobility for the a-ITO films is very high of around 50 cm2/Vs, which is 2-8 times higher than that of poly-crystalline ITO films prepared by conventional sputtering. The fabrication method reported here is promising for amorphous oxide films, especially for oxides whose amorphous phase is difficult to obtain.

Acknowledgments

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