Extended Abstracts of the 2018 International Conference on Solid State Devices and Materials, Tokyo, 2018, pp1291-1292

Growth mechanism of organic-inorganic hybrid perovskite thin films fabricated by two-step sequential vacuum evaporation method

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Abstract

We fabricated organic-inorganic hybrid perovskite (OHP) thin films, $CH_3NH_3PbI_3$ using the two-step sequential vacuum evaporation method (SVE) and found that the growth mechanism depends on the deposition rate of the PbI_2 layer in the first step. The deposition rates of PbI_2 were varied from 1 to 20 Å/s against the constant deposition rate (2 Å/s) of MAI for the second step. We characterized the formed films using x-ray diffraction, atomic force microscopy, UV-visible spectrometry, and x-ray photoelectron spectroscopy. A faster deposition rate of PbI_2 is better to form the well-defined perovskite structure. The perovskite formation with a good quality is assumed to depend on the density and roughness of PbI_2 in the first stage of the SVE method.

1. Introduction

Recently, organic-inorganic hybrid perovskites (OHP), described by ABX_3 (A = organic cation, B = metal cation, X = halogen anion) has attracted much attention as a new solar-cell material[1,2] with easy-fabrication, low-cost, and high-efficiency. OHP was reported to have a large absorption coefficient (direct bandgap of ~1.55 eV) and high mobility for electrons (7.5 $cm^2/V{\cdot}s$) and holes (12.5 $cm^2/V{\cdot}s$). The power conversion efficiency in the laboratory scale solar-cells is increasing rapidly for 5 years and 22.7 % now. Therefore, OHP materials are motivated not only to enhance performance for solar-cell application but also to bring to light new phenomena relevant to other technological application such as light emitting diode, laser, optoelectronics and thermos electronics, among others [3]. On the other hand, OHP has the disadvantage of low stability occurred by exposing water and heat. Unfortunately, however, several radical suggestions to solve the instability of OHP have not been obtained at present[4]. Normally, many researchers use the solution-prepared methods that has a weak crystallinity by the oxygen-incorporated contaminations from a remained solvent. On the other hand, the vacuum-evaporation method has a merit of contamination-free. Recently, M.-C. Jung, *et al*. have suggested a two-step sequential vacuum evaporation (SVE) method (Fig. 1) for Sn-based perovskite thin film[5]. However, there is no result of Pb-based perovskite thin film.

In this study, we performed the SVE method for the fabrication of $CH_3NH_3PbI_3$ (MAPI) with the first step for PbI_2 and the second step for MAI. And the formed films with different deposition rates on the fabrication of first PbI_2 layer were characterized by x-ray diffraction (XRD), atomic force microscopy (AFM), uv-visible spectrometer (UV-Vis.), and x-ray photoelectron spectroscopy (XPS). Finally, we discuss the growth mechanism in the SVE method for the fabrication of OHP thin film.

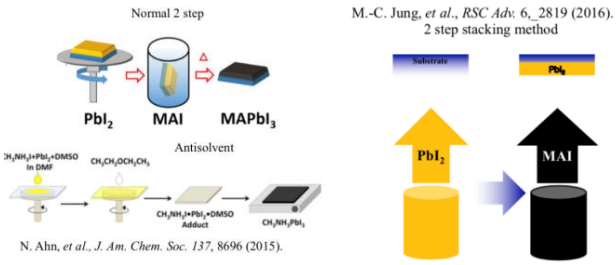

Figure 1. Simple schematic of the normal 2-step solution, antisolvent, and 2-step sequential vacuum evaporation methods.

2. Experiment

OHP thin films were fabricated by the SVE method in the customized vacuum chamber. A silicon, glass or sapphire substrates was cleaned by sonication in acetone for 10 minutes, rinsed in heated acetone, and UV-Ozone treatment just before loading into a vacuum chamber. A PbI_2 was deposited onto the substrates at room temperature in a vacuum (8.0×10^{-3} Pa). The deposition rate was maintained to 1.0, 3.0, 6.0, 10 and 20 Å/s and the film thickness was 100 nm, which was confirmed by a thickness monitor sensor. Then, the CH_3NH_3I (MAI) was deposited onto the PbI_2 with the deposition rate of 2.0 Å/s and the film thickness of 400 nm.

The crystal structure of formed samples were assigned by the XRD system in θ-2θ geometry with a Cu$K\alpha$ (1.54 eV) source. The surface morphology and chemical state were obtained by AFM (RINT-TTRIII, Rigaku) and XPS

(PHI5000 Versa ProbeII, ULVAC-PHI) with the monochromated Al*K*α source. To confirm the optical bandgap of formed perovskite, we used a UV-Vis spectrometer (V-770, JASCO).

3. Results and Discussion

In the different deposition rate of PbI_2, we can observe the different formation. (Fig. 2) Only the 10 Å/s sample of the PbI_2 deposition rate can form the perovskite with the brownish color. The surface roughness (RMS) is quite different.

In the presentation, we will discuss about the growth mechanism in the SVE method with all results of AFM, XRD, UV-Vis, and XPS measurements.

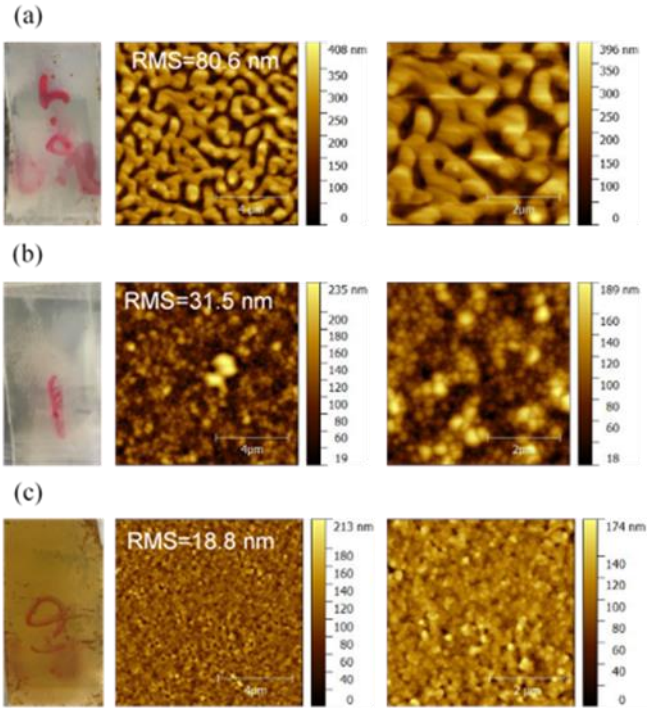

Figure 2. The surface morphologies with (a) 0.2, (b) 1, and (c) 10 Å/s of the PbI_2 deposition rate. Only the 10 Å/s sample of the PbI_2 deposition rate can form the perovskite with the brownish color.

4. Conclusions

We performed and characterized the fabrication of OHP thin film using the SVE method and AFM/XRD/UV-Vis/XPS, respectively. In the SVE method for OHP thin film, the perovskite growth depends on the PbI_2 deposition rate. From our approach, we expect to make a well-defined OHP thin film with no contamination.

Acknowledgements

This work was supported by funding from JSPS KAKENHI Grant No. 17K05033 (Japan) and Murata Science Foundation.

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