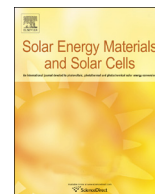




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journal homepage: www.elsevier.com/locate/solmatControlled crystallization of $\text{CH}_3\text{NH}_3\text{PbI}_3$ films for perovskite solar cells by various $\text{PbI}_2(\text{X})$ complexesWeifei Fu^{a,1}, Jieli Yan^{a,1}, Zhongqiang Zhang^a, Tao Ye^a, Yujing Liu^a, Jiake Wu^a, Jizhong Yao^b, Chang-Zhi Li^a, Hanying Li^{a,*}, Hongzheng Chen^{a,*}^a State Key Laboratory of Silicon Materials, MOE Key Laboratory of Macromolecular Synthesis and Functionalization, Department of Polymer Science and Engineering, Zhejiang University, Hangzhou 310027, China^b Microquanta Semiconductor Ltd., Hangzhou, China

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ABSTRACT

Lead halide complexes are efficient precursors to obtain high quality perovskite films and high performance solar cells. However, how the lead halide complex affects the crystallization kinetics and thus the film quality of perovskite has not been systematically studied. Herein, two $\text{PbI}_2(\text{X})$ complexes with different coordination strength of ligand X in complexation with PbI_2 ($\text{PbI}_2(\text{DMF})$ and $\text{PbI}_2(\text{DMSO})$) are synthesized. The correlation between the morphology, evolution of the perovskite formation, defects state, carrier lifetime and the subsequent photovoltaic performance are investigated in details via a two-step coating method with various $\text{PbI}_2(\text{X})$ complexes. We find that $\text{PbI}_2(\text{DMSO})$ derived perovskite film shows larger grains and vertically oriented grain boundaries as well as enhanced photoluminescence intensity and longer carrier lifetime. As a result, highly efficient inverted planar perovskite solar cells with a power conversion efficiency up to 17.0% are achieved from $\text{PbI}_2(\text{DMSO})$ complex under 1 sun illumination. This research could open up a new pathway to further improve the performance of perovskite solar cells through the control of perovskite crystallization kinetics by choosing proper $\text{PbI}_2(\text{X})$ complex precursors.

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1. Introduction

Perovskite solar cells (PVSCs) are a promising next-generation technology to produce lightweight, low-cost and flexible solar cell panels [1–12]. Currently, power conversion efficiencies (PCE) over 20% have been achieved for PVSCs, highlighting the exceptional photovoltaic properties of perovskite materials [13,14]. The photovoltaic properties of the perovskite films are greatly dependent on the film quality (such as coverage, morphology, grain size, grain boundary and so on) and thus the crystallization process of perovskite [15–19]. Accordingly, efforts have been made to examine the crystallization kinetics. Firstly, the basic film processing parameters have been investigated by tuning the annealing temperature [20–23] and time [21], spin-coating rate [24], the atmospheric conditions [25,26], or by employing different solvents [22,27,28] or additives [29–33]. Secondly, the starting materials including inorganic lead source and organic ammonia salt have been found to determine the perovskite crystal growth kinetics, which in turn

affects the film morphology and device performance [34–40]. Recently, a stable complex of methyl ammonia iodide or bromide ($\text{MAI}(\text{Br})$), lead iodide or bromide ($\text{PbI}(\text{Br})_2$) and dimethyl sulfide (DMSO) was demonstrated to be a decisive factor in retarding the rapid reaction between $\text{MAI}(\text{Br})$ and $\text{PbI}(\text{Br})_2$ in a solvent engineering method, which enabled the formation of a highly uniform and dense film [41–43]. Very recently, high quality formamidinium lead iodide (FAPbI_3) perovskite film was fabricated via the reaction between FAI and $\text{PbI}_2(\text{DMSO})$, providing an effective protocol for fabricating efficient PVSCs [13]. Later, the self-assembly of $\text{PbI}_2(\text{DMSO})$ complex was demonstrated to benefit the formation of ultraflat and dense perovskite film [44]. Complexes of PbI_2 and hydrogen halide are another type of efficient precursors. $\text{PbI}_2(\text{HI})$ was used to fabricate highly uniform FAPbI_3 films through a one-step spin-coating process [45]. Whereas $\text{PbI}_2(\text{HCl})$ complex was demonstrated to accelerate the formation of a pinhole free planar MAPbI_3 perovskite film in both one-step and sequential deposition methods [46]. These results indicate that lead halide complexes are efficient precursors to obtain high quality perovskite films and thus high performance solar cells.

Nevertheless, how can the lead halide precursor affect the perovskite formation which further governs its crystallization kinetics to determine the overall film quality has not been

* Corresponding authors.

E-mail addresses: hanying_li@zju.edu.cn (H. Li), hzchen@zju.edu.cn (H. Chen).¹ These authors contributed equally to this work.

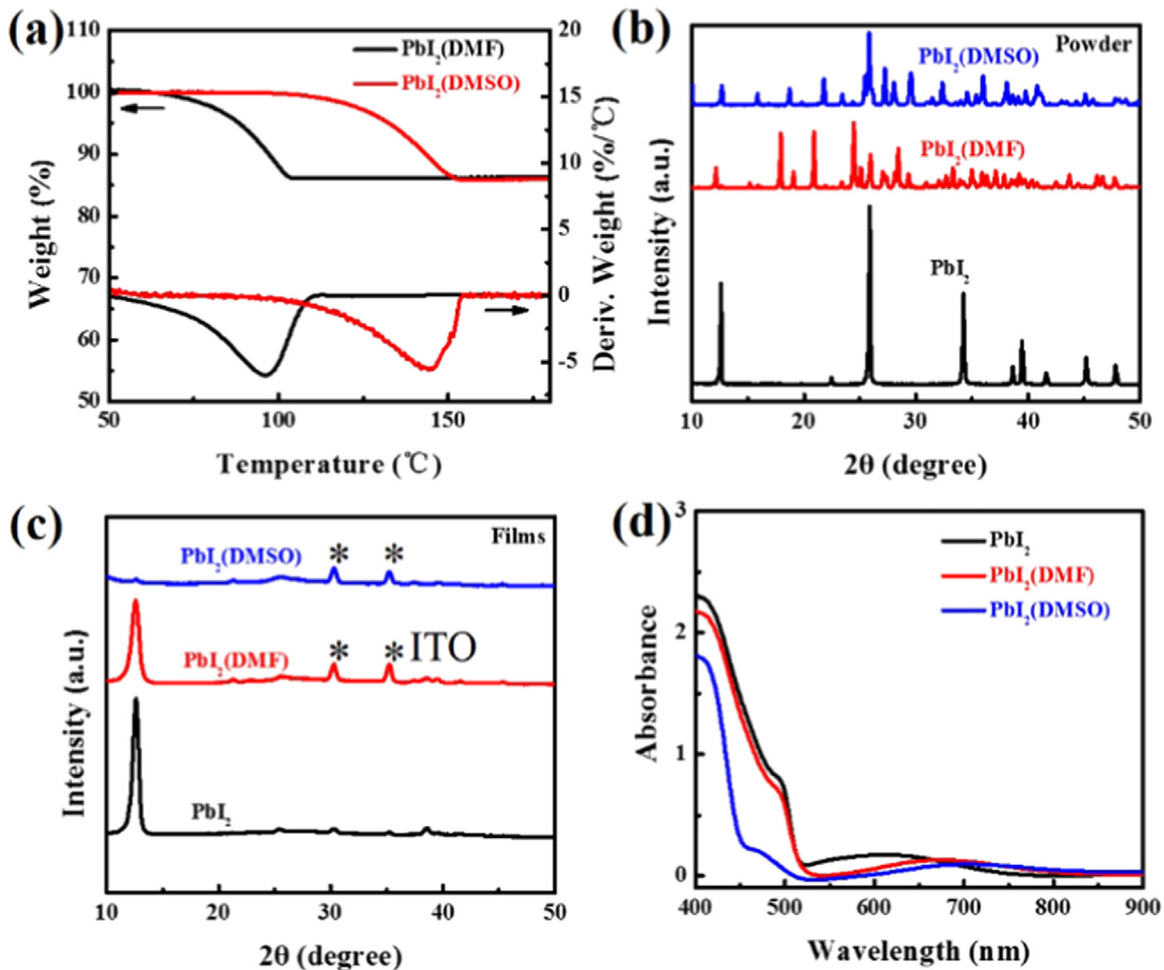


Fig. 1. (a) TGA curves of PbI₂(DMF) and PbI₂(DMSO). XRD patterns of PbI₂ and PbI₂(X) powders (b) and their as-prepared films (c). (d) Absorption spectra of PbI₂ and PbI₂(X) films.

systematically studied yet. To elucidate a clear correlation of this transformation could contribute to develop efficient solar cells derived from high quality perovskite absorber layers, herein, we explore the ease fabrication of crystalline perovskite films from two synthesized PbI₂(X) precursors, PbI₂(DMF) and PbI₂(DMSO) via a simple two-step coating method. Through studying the perovskite formation over different precursors, crystallization kinetics and film qualities, carrier lifetimes, defects etc., we reveal that the presence of solvent molecule as ligands in lead halide precursors, like DMF and DMSO, is effective to promote the conversion of precursors to perovskite for high-quality crystalline film formation. For instance, PbI₂(DMF) with the weak coordinating ligand (DMF) allow being quickly converted into MAPbI₃ perovskite even at room temperature, whereas strong ligand DMSO constituted PbI₂(DMSO) requires a thermal annealing to facilitate the conversion process. These indeed provide means to further control the crystallization kinetics of perovskite film. The resultant perovskite film made from PbI₂(DMSO) precursor showed densely packed large grains, of which the grain boundaries are vertically aligned, leading to a much reduced film defects and longer carrier lifetime than those from PbI₂(DMF) and pristine PbI₂. As a result, highly efficient planar PVSCs with a PCE up to 17.0% have been achieved from PbI₂(DMSO) precursor.

2. Experimental

2.1. Materials

Unless stated otherwise, all materials were purchased from Sigma-Aldrich and used as received. PC₇₁BM was purchased from American Dyes Source, Inc. CH₃NH₃I (MAI) was purchased from Shanghai Materwin New Materials Co. Ltd. Poly(bis(4-phenyl)(2,4,6-trimethylphenyl)amine) (PTAA) was purchased from Xi'an Polymer Light Technology Corporation. PbI₂(DMSO) was synthesized according to the reported procedure [13]. In a typical synthetic process, PbI₂ (50 g) was dissolved in dimethylsulfoxide (DMSO, 150 mL) at 60 °C and then toluene (350 mL) was slowly added into the PbI₂ solution. The produced white precipitation was filtered and dried for 3 h at room temperature and then annealed for 24 h in vacuum oven at 60 °C. PbI₂(DMF) was synthesized in a similar procedure. PbI₂ (50 g) was dissolved in *N,N*-dimethylformamide (DMF, 150 mL) at 60 °C and then toluene (350 mL) was slowly added into the PbI₂ solution. The produced light yellow precipitation was filtered and dried for 3 h at room temperature. ZnO nanoparticles were synthesized by a sol-gel process using Zn acetate and tetramethylammonium hydroxide (TMAH) [47]. A stoichiometric amount of TMAH dissolved in ethanol (0.55 M) was added dropwise to Zn acetate dihydrate

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