

CO₂ laser-assisted ultrafast crystallization for highly efficient perovskite solar cells

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Introduction

Perovskite solar cells have achieved tremendous progress since the first report in 2009, with power conversion efficiencies already comparable to their silicon counterpart nowadays.^{1,2} Crystallinity controlling has always been the central focus over the development history. Numerous efforts including composition exploration, device structure optimization, process improvement, as well as passivation strategies, have been dedicated to enhance crystallinity and thus improving device performance. Heat treatment is the most critical factor in perovskite crystallization process, which provides activation energy for phase transition. There are multiple heating strategies being utilized nowadays, including traditional hot-plate thermal annealing (TA), microwave annealing (MA) and laser annealing (LA). Among them, traditional thermal annealing is the most extensively employed in literature reports. Laser annealing possesses various competitive advantages towards thermal annealing, such as non-contact annealing, highly localized or selective annealing with much less adverse thermal effects on adjacent functional layers and the substrate. This is beneficial for heat-sensitive substrates. Some research groups have reported laser annealing for perovskite solar cells with promising results.^{3,4,5} For most of these reports, UV lasers, visible wavelength lasers, and near infrared lasers, they have short wavelengths which means high energy that can be detrimental to chemical bonds in the perovskite photo-absorber [5]. They also use short pulse widths (nanosecond to femtosecond) which can be damaging to the charge transport layers [6], adjacent to the perovskite absorber. CO₂ lasers, on the other hand, produce much longer wavelengths (9.3 - 10.6 μm) and thus lower photon energy, which are less damaging to perovskites.

Herein, we have successfully developed a far infrared (FIR) carbon dioxide (CO₂) laser process for FAPbI₃ perovskite crystallisation under ambient condition. Details of the laser parameters will be presented in the conference. As shown in **Figure 1**, crystallization process is most affected by laser power density. We also compared ambient thermal annealing (ATA) with ambient laser annealing (ALA) on the performance of ITO/MeO-2PACz/ FAPbI₃ /C60/BCP/Cu solar cells. Results (**Table 1**) show the advantage of ALA producing cells with better performance. The champion cell produced a power conversion efficiency (PCE) of 21.8%, higher than the efficiency, at 17.4%, of the ATA cell. This result is respectable for a laser annealed MA-free pristine perovskite solar cell without any additive, passivation or post treatment.

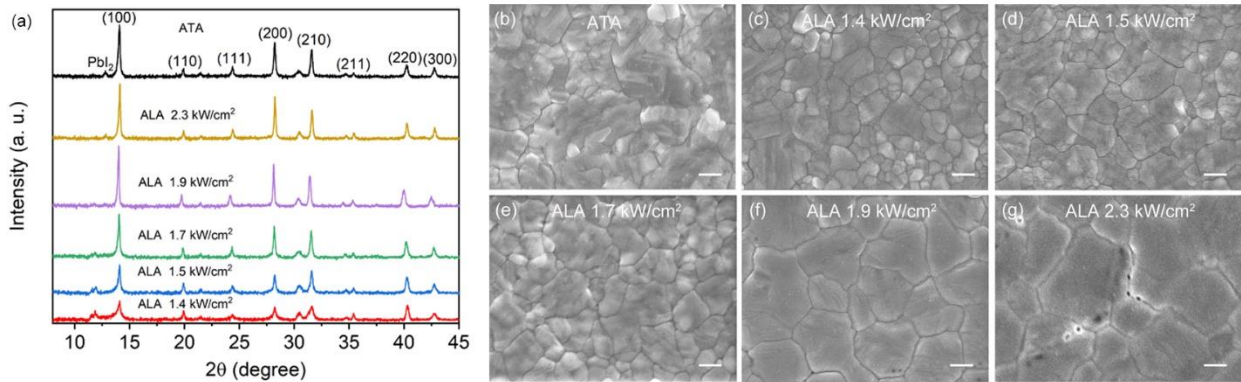


Figure 1. (a) X-ray Diffraction (XRD) patterns and top view scanning electron microscopy (SEM) images of (b) ATA and (c-g) ALA films using laser power densities (c) 1.4 kW/cm², (d) 1.5 kW/cm², (e) 1.7 kW/cm², (f) 1.9 kW/cm², (g) 2.3 kW/cm². Scale bar, 300 nm.

Table 1. Electrical characteristics of perovskite solar cells using ambient thermal annealing (ATA) or ambient laser annealing (ALA).

Annealing method	$J_{SC}/\text{mA cm}^{-2}$	V_{OC}/mV	FF/%	PCE/%	$J_{int.}/\text{mA cm}^{-2}$
ATA	23.9	980	75.2	17.6	23.5
ALA	24.8	1058	82.9	21.8	24.7

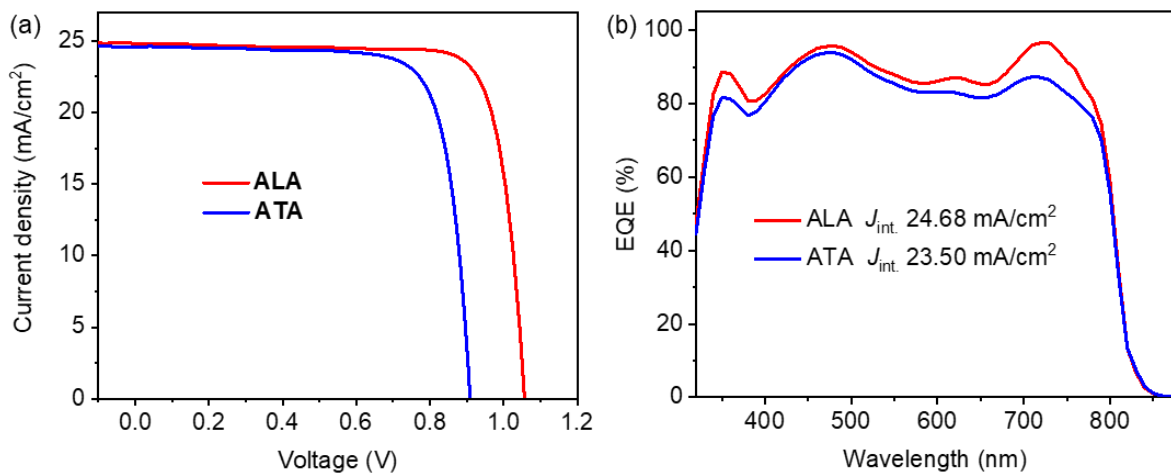


Figure 2. (a) Current density vs voltage (J - V) and (b) external quantum efficiency (EQE) of perovskite solar cells using ATA and ALA.

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