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Plasma characteristics of 355 nm and 532 nm laser deposition of Al-doped ZnO films

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ABSTRACT

The plasma characteristics of 355 nm and 532 nm laser ablation of Al-doped ZnO (AZO) target were studied by optical emission spectroscopy and ion probe measurements. Zn emission lines were measured detected at above 0.9 J/cm^2 for 355 nm laser and 0.6 J/cm^2 532 nm laser respectively, while Al species were detected only above 2 J/cm^2. The kinetic energy of the ions was slightly higher for 532 nm ablation as compared to 355 nm ablation. In addition, the ablation of 532 nm laser was affected by the large laser penetration depth. When deposited at 2 and 4 J/cm^2, AZO films with energy band gap of 3.45–3.6 eV were obtained. Nanostructured AZO films were obtained by 355 nm laser ablation but nano and micro-particulates were formed in 532 nm laser ablation. The large micron-sized particulates were Al-rich thus affecting not only the morphology but also the stoichiometry of the films. It is thus concluded that despite a lower ablation and growth rate, 355 nm generated Zn, O and also Al species at lower threshold fluence that can lead to high quality AZO films at room temperature.

1. Introduction

Pulsed laser deposition has been a versatile method for deposition of single and multi-elements coating and films, nanomaterials, metastable materials and some that are not feasible to be obtained by other methods. Clean photons from the laser ablate a target material to produce plasma plume of neutrals, ions and molecules. These species travels with specific kinetic energy that leads to nanomaterials growth on the substrate where the optical and electrical properties are governed by the size and morphology. The method has been successful in growing various ZnO-based films and nanostructures [1–10]. In general, for every laser wavelength and pulse length, threshold fluence is needed for ablation while deposition in O_2 background gas enables stoichiometry ZnO films. Substrates are typically heated to improve crystallinity of the films but recently it was found that under specific condition, low substrate temperature/room temperature can be employed to grow nanocrystalline ZnO films on plastics substrates [11–14]. The findings inferred that materials growth is solely contributed by the energetic plasma plume, without additional heating that increase the adatom mobility. However, the plasma characteristics that are responsible for the growth have not been widely explored and the correlation between plasma plume and ZnO properties have only been reported by a few research groups. The effects of ions energy by ns and fs laser on the growth of ZnO films were identified in experiment and plasma dynamic simulation [15]. Ns laser can produce ions of up to a few hundred eV while fs laser ablation produce ions up to KeV range. The bombardments of highly energetic ions induced stress and defects in the films while moderately energetic species promote atomic surface diffusion for ordered materials growth. It has also been shown that the ZnO films properties were dependent on the substrate position [14]; possibly due to exposure to the different localized region in the plasma plume. In another report, 193 nm laser produced plasma of ZnO was shown to consist of fast ions and slow neutrals, observed by using wavelength and spatially resolved optical emission spectroscopy and Langmuir probe. The slow component was contributed by materials redeposited on the target and this would result in a non-stoichiometric deposition [16]. Subsequently, the optical emission spectra analysis of 355 nm laser-produced plasma from ZnO suggested that film deposition should be performed in O_2 at a substrate-target distance beyond 10 mm to achieve the optimum electron temperature (T_e) and density (n_e) [17]. On the other hand, it was reported recently that AZO films can be grown without substrate heating by utilizing dual-confined plasma [18,19], where the temperature of the substrate rises to 50–150 °C because of the plasma process; depending on the operating power and background pressure.

In this work, the plasma species and ion energies in pulsed laser ablation of an Al-doped ZnO (AZO) target by 355 nm and 532 nm laser were measured by using time-integrated optical emission spectroscopy and ion probe measurement. Governed by both properties of the laser and the zinc oxide target, the degrees of absorption, heating, ablation were responsible for the plasma formation. The penetration depth in

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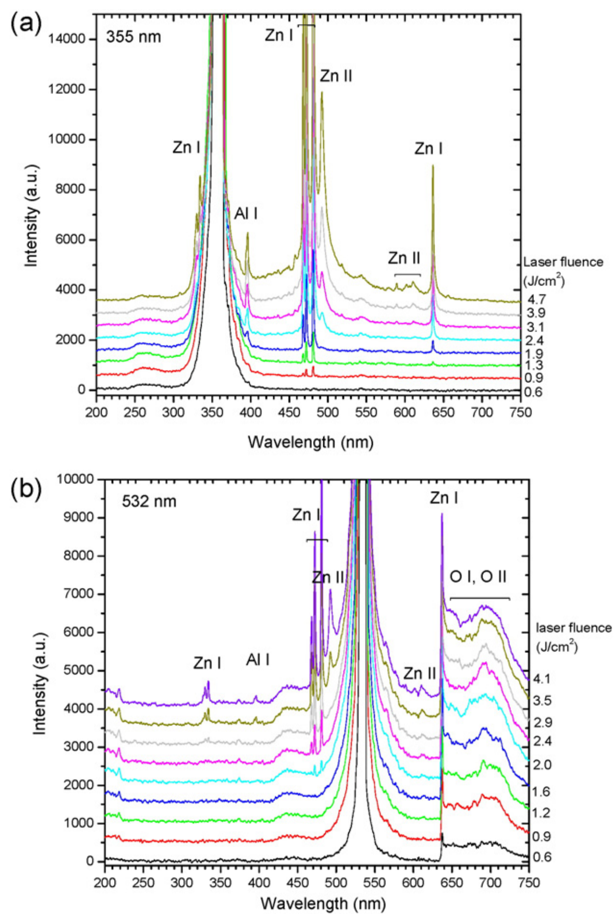

Fig. 1. Optical emission spectra of ablation of AZO target by (a) 355 nm and (b) 532 nm laser. Zn I (472.21 nm and 481.05 nm) was detected at 0.9 J/cm^2 while Zn I (637 nm) was detected at ~0.6 J/cm^2 for 532 nm ablation.

laser ablations depends on thermal diffusion distance (T) of the material and optical absorption length (A) of the laser pulse: $T = (2D\tau)^{1/2}$ and the absorption length of laser in target material $A = (1/\alpha_t)$ where α_t = wavelength dependence absorption coefficient, D = thermal diffusivity, τ = pulse length. Thus, for ns laser ablation of the same materials,

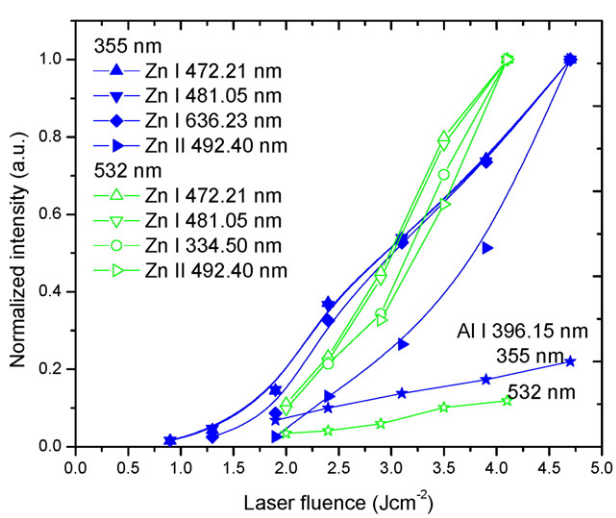

Fig. 2. The normalized intensity of the characteristics lines: Zn I (334.50 nm, 472.21 nm, 481.05 nm, 636.23 nm), Zn II (492.40 nm) and Al I (396.15 nm) are plotted against laser fluence. Zn I 636.23 nm for 532 nm ablation, although detected at 0.6 J/cm^2 was not compared as it overlapped with other emission lines. The emissions from Zn II and Al I only occurred at and above 2 J/cm^2.

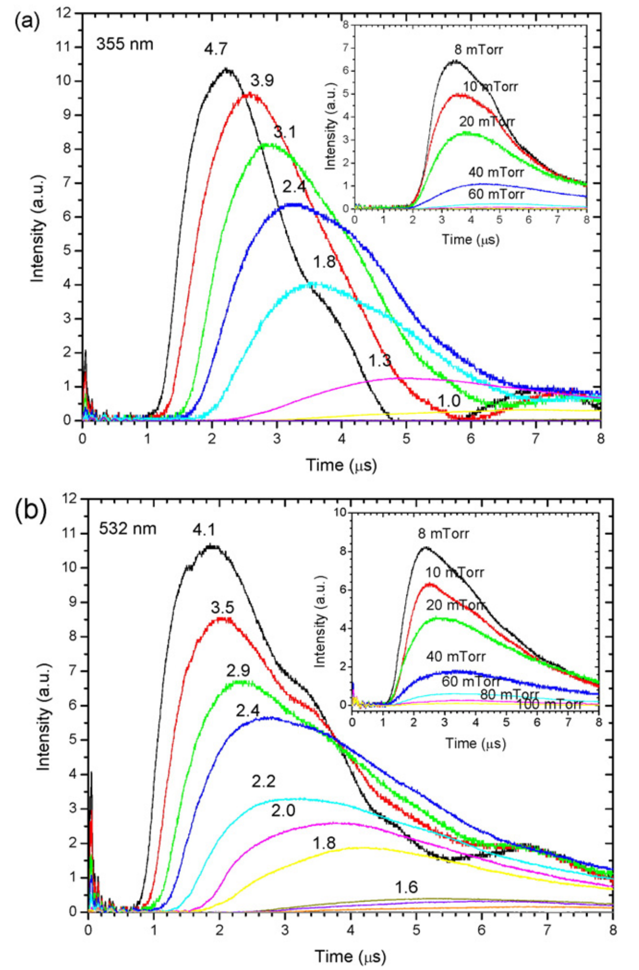

Fig. 3. Time of flight signals of (a) 355 nm and (b) 532 nm laser ablation of AZO target at different laser fluence. Insets show the ions signal measured at 2 J/cm^2 at different O_2 background pressure.

the difference of laser wavelength led to the difference in absorption coefficient. The absorption coefficient of ZnO is higher at 355 nm as compared to 532 nm [20,21], which results in the lower penetration depth for 355 nm laser. The ablation process by both lasers was performed to grow Al-doped ZnO films at fixed background gas and at room temperature. The work aimed to explore the ablation process and the plasma characteristics by 355 nm and 532 nm laser, in relation to the properties of AZO film.

2. Experimental

A Nd-YAG laser (second and third harmonic, 532 nm and 355 nm, 4.7 ns) (EKSPLA, NL301) was used for ablation of Al doped ZnO target (Kurt J. Lesker, 99.99% purity, 98% ZnO + 2 wt.% Al_2O_3). The deposition chamber was first evacuated to high vacuum of 10^{-6} Torr, and then introduced with O_2 gas to obtain a partial pressure of 16–20 mTorr for thin film deposition. Silicon (p-Si 100) and glass (Corning #26003) substrates were ultrasonically cleaned prior to use. They were placed 5 cm from the target.

Deposition of AZO films was carried out at room temperature for 90 min at a repetition rate of 10 Hz. The laser beam was focused into an elliptical spot on the target at 45° with the dimension of ~1 mm × 0.8 mm. The laser energy was controlled to produce laser fluences of 0.6 to 4.7 J/cm^2. The laser beam raster an area of 6 mm^2 on the target surface. No post-growth annealing was performed. The plasma plume was measured by using time-integrated optical emission spectroscopy where the emission was imaged onto a quartz fiber