

Investigation on structural and optoelectronic properties of in-situ post growth annealed ZnSnN₂ thin films

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ABSTRACT

Zinc Tin Nitride (ZnSnN₂) has been proposed as a new potential solar absorber for energy applications since its properties are similar to group III nitrides. But the main challenging issue for this material is to reliably synthesize films with carrier density 10¹⁷ to 10¹⁸ cm⁻³. In this regard, ZnSnN₂ thin films were successfully developed by radio frequency (RF) magnetron sputtering and a systematic in-situ post growth annealing was performed in order to study its effect on the optoelectronic properties. At 500 °C, in-situ post growth annealed film possess good crystalline nature (69 nm) and also exhibits low carrier concentration (−3.97 and −2.42 × 10¹⁸ cm⁻³) and high mobility (14.5 and 11.1 cm² V⁻¹ s⁻¹). The optical bandgap of the annealed ZnSnN₂ film is in the range of 1.78–1.71 eV. Therefore, annealed ZnSnN₂ can be regarded as potential solar absorber and alternate material for III-V nitrides.

1. Introduction

Group (III) nitride semiconductors such as GaN and InN have been widely applied in the field of optoelectronics in the past few decades, due to their unique properties and suitable band gaps in the range of 1.0–3.5 eV [1]. However, the rarity of Ga and In rather than the cost have led researchers to find new novel materials. Recently, II-IV-N₂ semiconductor materials are widely regarded as favorable materials for replacing group III-Nitrides based on theoretical calculations as well as experimental results [2–4]. In particular, Zn-IV-N₂ semiconductor materials have been attracting a lot of attention towards potential earth abundant material to be used in optoelectronic devices and serves as an alternative to InN and GaN. The replacement of III element in III-nitride analog with Zn and Sn cations in an ordered manner lead to the formation of ZnSnN₂ with stoichiometric ratio of 1:1:2 (Zn:Sn:N) [5].

ZnSnN₂ is predicted to crystallize in the orthorhombic crystal structure, which is derived from the wurtzite lattice that is the characteristic of III-nitride semiconductors. ZnSnN₂ possesses useful properties such as direct band gap and relatively low effective masses for both electrons and holes as compared to the other Zn-IV-N₂ materials and hence acts as effective material for optoelectronic applications, particularly solar cells. Recently, Chen et al. [2] proposed ZnSnN₂ as suitable solar cell absorber material. In spite of these significant qualities, ZnSnN₂ is one of the least studied member of the Zn-IV-N₂ class,

due to the high carrier density (10²⁰ cm⁻³) which in turn leads to the non-rectifying behavior in a diode and short minority carrier lifetime and complication in the optical bandgap analysis (2 eV). The growth of ZnSnN₂ with enhanced structural and optoelectronic behavior is also a challenging task to the researchers.

To date, only a few studies have been made on reducing the carrier density of ZnSnN₂ by growing or by subjecting to post growth annealing [6]. Recently Deng et al. [7] reported the carrier concentration of 1.6 × 10¹⁸ cm⁻³ and the maximum mobility of 3.98 cm² V⁻¹ s⁻¹ for annealed ZnSnN₂. In our present work, ZnSnN₂ have been synthesized by RF magnetron sputtering and performed in-situ post growth annealing to study its effect on carrier concentration and mobility. The phase structure was found to be orthorhombic with preferential growth along (002) orientation. Additionally, the compositional analysis of ZnSnN₂ films was also studied and presented the results. Based on the observation, it was found that the in-situ post growth annealing induced the better crystalline nature with low carrier density and high mobility, indicating the potential use of annealed ZnSnN₂ films for photovoltaic applications.

2. Experimental section

The deposition of ZnSnN₂ thin films was recently reported by our group using reactive RF magnetron sputtering (Fig. 1(a)) [8]. The RF

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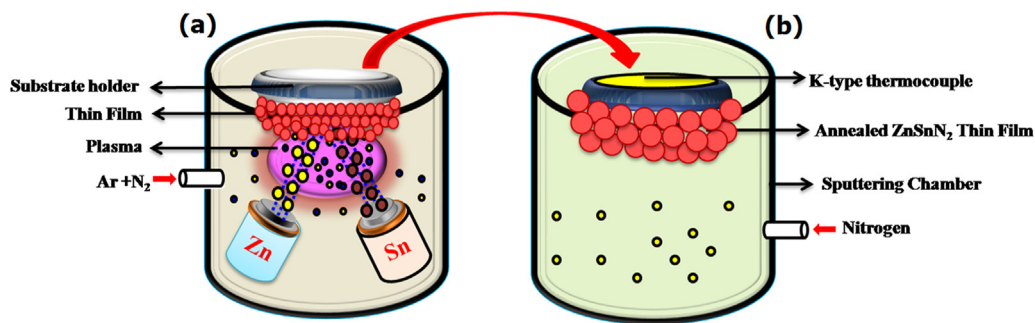


Fig. 1. (a) Experimental set up for reactive RF magnetron co-sputtering and (b) In-situ post growth annealing process of ZnSnN₂ thin films.

power was 40 (± 1) W for Zn and 10 (± 1) and 15 (± 1) W for Sn and the films were deposited for 1 h at 450 (± 5) °C substrate temperature. After the deposition, films were subjected to in-situ post growth annealing at 500 (± 5) °C for 3 h under nitrogen atmosphere at a pressure of 12 mbar in the same vacuum chamber, in which, the films were deposited (Fig. 1(b)).

3. Results and discussion

Fig. 2(A) shows the X-ray diffraction (XRD) patterns of as-grown and in-situ post grown ZnSnN₂. From Fig. 2(A), we inferred that ZnSnN₂ crystallize in orthorhombic system. Six distinct peaks were observed in the as-deposited ZnSnN₂ film, revealing its crystalline nature and the calculated lattice parameters are consistent with orthorhombic ZnSnN₂ (Table 1). It is noteworthy that Table 1 compares the calculated and experimental lattice parameters of orthorhombic ZnSnN₂ reported by

Table 1
Comparison of calculated and observed lattice parameters for orthorhombic phase structure of ZnSnN₂.

Lattice parameters	Orthorhombic		Experimental (Å) Orthorhombic (In this work)	
	Calculated (Å) [11]	Experimental (Å) [7]	As-Deposited	Annealed
a	5.85	5.85	5.79	5.79
b	6.76	6.74	6.74	6.72
c	5.58	5.49	5.59	5.45

Punya et al. [11] and deng et al. [7] to the lattice parameters observed in this work. It was also observed that the peaks were shifted towards higher angle for in-situ post growth annealed films (Fig. 2(A)). Interestingly, the crystallite size of in-situ post grown ZnSnN₂ thin films

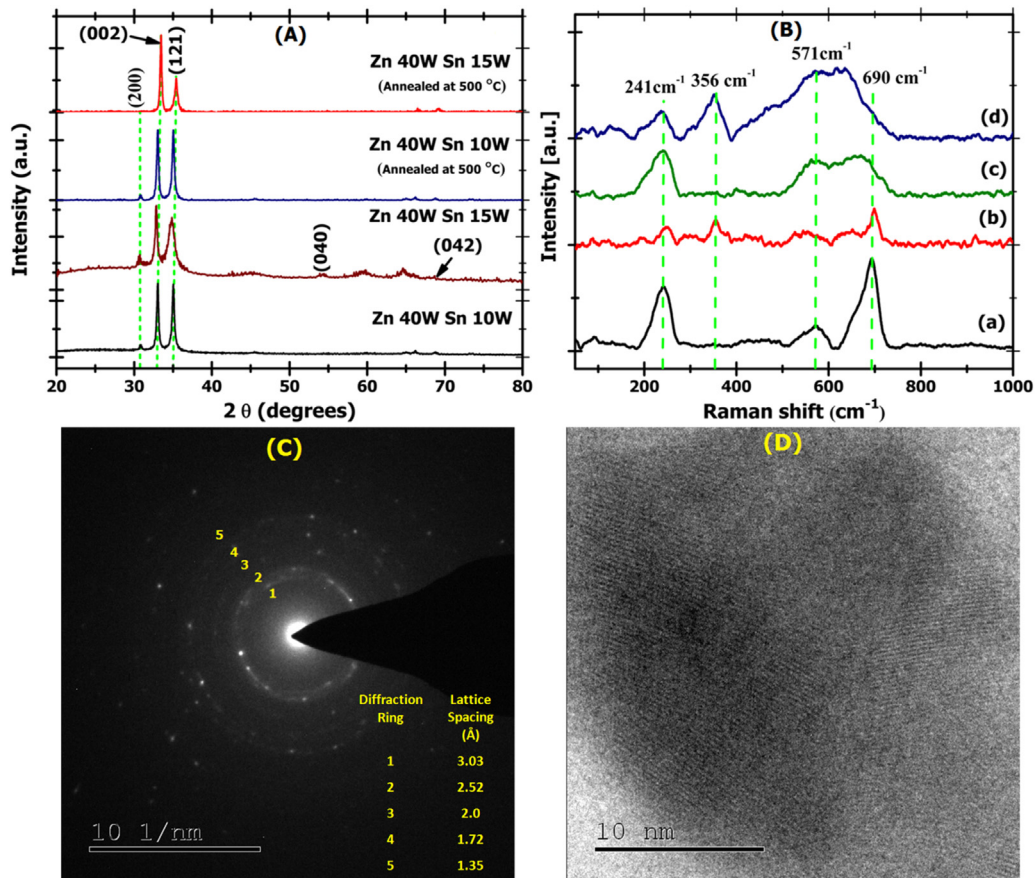


Fig. 2. (A) XRD patterns and (B) Raman spectra of as-deposited and in-situ post growth annealed ZnSnN₂ thin films. (C) TED pattern and (D) Top-Down HRTEM images of post growth annealed ZnSnN₂ thin films.

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