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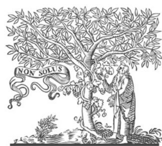

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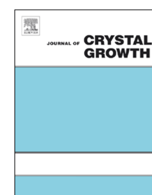

Pulsed laser deposition: A viable route for the growth of aluminum antimonide film

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ABSTRACT

Aluminum antimonide films (AlSb) were successfully deposited on glass substrates by ablating an aluminum antimonide target using pulsed Nd-YAG laser. Films deposited at substrate temperatures ~773 K and above showed zinc blende structure. Increase in substrate temperature culminated in grain growth in the films. Photoluminescence studies indicated a strong peak ~725 nm (~1.71 eV) and ~803 nm (~1.55 eV). Films deposited at higher deposition temperatures indicated lower residual strain. Characteristic Raman peaks for AlSb at ~151 cm^{-1} followed by two peaks located at ~71 cm^{-1} and ~116 cm^{-1} were also observed.

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1. Introduction

AlSb, a III–V binary compound semiconductor, with indirect band gap of 1.62 eV, is ideal for solar spectrum absorption [1]. It has a high theoretical photoelectric conversion efficiency of ~27% [2]. Potentiality of AlSb to be used in high energy detector technology has been suggested by Yu et al. [3]. AlSb films were utilized as a buffer layer for growing InSb and GaSb on different substrates [4–8]. Aluminum has a shorter diffusion length compared to that of In and Ga. This facilitates the formation of a closed AlSb layer at an earlier stage of growth [9–12] leading to the formation of InSb and GaSb compounds.

The constituent elemental materials are relatively abundant, non-toxic and low cost making AlSb attractive for device applications [13,14]. Despite the above advantageous properties, ease of availability and possible applications, the reports on AlSb material are limited.

In the past, coevaporation, hot wall epitaxy and co-sputtering techniques were adopted for the preparation of AlSb thin films. Deposition of AlSb films by coevaporating elemental Al and Sb onto glass substrates kept at 550 °C was reported by Johnson [15]. Lal et al. [16] carried out investigations on the microstructural features of AlSb thin films grown by thermal evaporation under various conditions. Bulk AlSb was used as a source for the thermal evaporation and AlSb films were deposited on various substrates at different temperatures. Polycrystalline thin films of AlSb deposited by co-sputtering were reported by Hao et al. [17]. The structure, morphology and electrical properties of the films were studied. The AlSb films had face-centered cubic structure. It was observed that use of hydrogen in the ambience and deposition at higher substrate temperature was beneficial to the grain growth. The molecular beam epitaxy technique was adopted by Proessdorf et al. [18] to study the growth of AlSb thin films. The structure, morphology and electrical properties of AlSb films under different deposition conditions were investigated by them. Dhakal et al. [19] synthesized AlSb thin film by co-sputtering Al and Sb targets and fabricated p–n and p–*i*–n device structures. Photovoltaic response of the devices was also investigated by them. The MBE technique was adopted by Mori et al. [20] to deposit AlSb layers. These layers were used as the buffer for the formation of heteroepitaxial InSb films on Si (001). Except MBE technique, majority of the techniques used by previous workers to deposit AlSb films encountered difficulties to ensure compositional reproducibility.

We report here the synthesis of AlSb films deposited on glass substrates by utilizing the PLD technique. The films are characterized by X-ray diffraction (XRD) and field emission scanning electron microscope (FESEM) for microstructural studies. Photoluminescence (PL), Raman and X-ray photoelectron spectroscopy

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(XPS) techniques were also utilized for optical and bonding environmental studies.

2. Experimental details

The deposition of aluminum antimonide films has been carried out in a conventional PLD configuration. It consisted of a multi-port stainless steel chamber and a laser system. The cylindrical vacuum chamber is equipped with appropriate gas inlets, and a rotating target holder, capable of accommodating six targets. The substrates could be heated upto 1073 K by using ceramic heaters attached to the substrate holder. AlSb films were deposited by ablating a target made by admixing requisite amount of aluminum and antimony powders (both of 99.99% purity). A Nd:YAG laser (wavelength $\lambda=355$ nm, pulse duration $\tau=10$ ns, frequency $f=10$ Hz) was utilized to ablate the target to deposit films onto glass substrates kept at three different substrate temperatures (673 K, 773 K and 873 K). The laser beam was focused with a glass lens ($f=23$ cm) on the target at an angle of 45° with respect to the normal and the laser fluence was maintained at 8 J/cm^2. The rotation of the target was set at 10 rpm to avoid fast drilling. The distance between the target and the substrate was maintained at ~6.5 cm and the pressure during deposition was lower than 5×10^{-6} Torr. All the films were deposited for 5 min.

Energy Dispersive X-ray (EDAX) (Oxford Instruments' INCA Energy 250 Microanalysis System) analysis was utilized for compositional information. The surface morphology of the films was recorded by FESEM (Carl Zeiss SUPRA® 55 with GEMINI® Technology. This system has a resolution ~0.8 nm @ 15 kV). Rigaku MiniFlex XRD (0.154 nm Cu K_α line) was used to obtain the microstructural information. M/s SPECS (Germany) make spectrometer was used to carry out XPS measurements. Surface chemical properties of the films were obtained using the above system operated a pressure of ~4e−10 Torr. Al K_α line was used as the X-ray source at 1486.74 eV. Photoluminescence (PL) measurements were recorded at 300 K. A 300 W Xenon arc lamp was used as the emission source while a Hamamatsu photomultiplier along with a 1/4 m monochromator was used as the detecting system. Renishaw inVia micro-Raman spectrometer using 514 nm Argon laser was used to record the Raman spectra.

3. Results and discussion

Deposition of AlSb film by physical vapor deposition technique is indeed a tricky process. The elemental components have diverse properties, e.g., their melting point ($T_{m\text{-Al}}\sim660$ °C; $T_{m\text{-Sb}}\sim630$ °C) and the temperature at which the vapor pressure becomes $\sim10^{-4}$ Torr (for Al ~1010 °C; for Sb~425 °C). Moreover, Sb sublimes. The above properties debar single source evaporation and flash evaporation for depositing films with assured composition. Also, Al has a very low sputtering yield ~1.0 (at 500 eV argon ion energy) while Sb has a substantially high sputtering yield of ~2.83 (at 500 eV argon ion energy). Thus it would be extremely difficult to control the composition of AlSb films effectively by the sputtering technique. The above information evidently suggests the necessity of a technique which would permit emanation of the constituent materials in appropriate ratios to reach the substrate for forming films with assured composition. In this regard, the PLD technique has emerged as one of the most popular and effective techniques used in the present days for the deposition of stoichiometric thin films. In this technique, a pulsed laser is directed on a solid target. The nanosecond laser pulse is focused to give an energy density sufficient to vaporize a few hundred angstroms of surface material in the form of neutral or ionic atoms and molecules with kinetic energies of a few eV, which then get deposited onto the substrate. In PLD, the intensity of the laser is of the order of 10^8–10^9 W/cm^2 corresponding to pulse duration of a few nanoseconds. Therefore, there is enough time for the pulses to absorb, heat the target surface, and, finally, lead to the removal of matter. Additionally, the plasma temperature is high ($\sim10^3$ K) and the evaporants become more energetic when they pass through the plume. This affects the film deposition in a positive manner due to increase in the adatom surface mobility. The use of short pulses helps to maintain high laser power density in a small area of the target and produces congruent evaporation. The other advantage is the higher absorption coefficient of the laser pulse in the UV region so that the beam energy is absorbed on a thin surface layer and the ablation occurs more efficiently. The fluence of the laser pulse has to be larger than a certain threshold value so that all the species can be stoichiometrically removed from the target. The above advantageous properties of the PLD technique would be requisite for ablating AlSb from a target whose composition could be prefixed by taking into account the weighted average of the vapor pressures $\sim10^{-4}$ Torr of the elemental components constituting the film. It may be noted that we have used fresh targets for each deposition to avoid any plausible fractional distillation that might be occur despite all the measures taken.

Thus, the pulsed laser deposition (PLD) technique may be a viable alternative technique for the deposition of AlSb films with reproducible properties. It appears that so far, the PLD technique has not been employed for the deposition of AlSb films despite its advantageous property to grow complex multi-component materials with assured stoichiometry [21,22]. This technique ensures stoichiometric transfer of target material to the substrate and thus, would allow deposition of AlSb by ablating a target composed of two divergent components, as reported in this communication.

3.1. Microstructural studies

FESEM micrographs of AlSb films, deposited at 673 K, 773 K and 873 K for a fixed duration of 5 min on glass substrates are shown in Fig. 1a–c respectively. A typical EDAX spectrum for a representative AlSb films deposited at 773 K is represented in Fig. 1d. Grain sizes increased for films deposited at higher substrate temperatures (Table 1) Change in the surface texture of the AlSb films deposited at different substrate temperatures is indicated in Fig. 1a–c. The films became more compact when deposited at substrate temperatures greater than 673 K. The EDAX spectrum indicated the films to be slightly Sb rich (Table 1). But as the substrate temperature increased, the films became more stoichiometric in nature.

The X-ray diffraction (XRD) patterns for the above AlSb films are shown in Fig. 2. It was observed that AlSb films are polycrystalline in nature with compact grains. The films deposited at 673 K (curve-a) indicated amorphous nature. One may observe a broad hump located at $2\theta\sim45°$ for films deposited at 673 K (curve-a). However, the films deposited at temperatures ~773 K and above had predominantly zinc blende structure with peaks at $2\theta\sim28.9°$, $2\theta\sim41.8°$, and $2\theta\sim48.8°$. These corresponded to the reflections from planes of (200), (220) and (311) of AlSb (curves b and c) respectively [23]. During deposition, a higher substrate temperature would result in an increase in the adatom mobility which in turn would facilitate growth in the crystallite size. Thus, the improvement in crystallinity of the films when deposited at temperatures greater than 773 K would be expected. Stable zinc blende structure dominated the films. No peaks for Al or Sb are observed in the XRD trace which demonstrates that the films are only composed of AlSb. The lattice constant (0.610 nm), calculated from the XRD pattern, was in good agreement with that reported earlier for AlSb bulk (0.613 nm) with zinc blende structure [24].

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