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Characterizations of pulsed laser deposited SiC thin films

Y.S. Katharria [a,*], Sandeep Kumar [a], Ram Prakash [b], R.J. Choudhary [b], F. Singh [a], D.M. Phase [b], D. Kanjilal [a]

[a] *Inter-University Accelerator Centre, Aruna Asaf Ali Marg, P.O. Box 10502, New Delhi 110 067, India*
[b] *UGC-DAE Consortium for Scientific Research, Khandwa Road, Indore 452 017, India*

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Abstract

Thin films of silicon carbide (SiC) were prepared using pulsed laser deposition (PLD) on Si(100) substrates at a temperature of 370 °C. Various structural characterizations showed the development of short-range SiC precipitates in the films. These films were annealed isochronally at temperatures of 800 °C, 1000 °C and 1200 °C for 2 h under an inert environment. Thermally induced crystalline ordering of SiC into β-SiC phase was investigated by X-ray diffraction (XRD), Raman spectroscopy and Fourier transforms infrared (FTIR) spectroscopic measurements. In addition to the crystallization of SiC films, high temperature annealing resulted in the dissolution of carbon clusters found in the as-grown films.

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1. Introduction

A large set of promising electrical, optical and mechanical properties possessed by SiC makes it a very attractive candidate for high power, high temperature and high frequency applications even under the harsh environmental radiation and chemical conditions [1–5]. Among the various possible crystalline structures of SiC, also known as polytypes, cubic SiC phase (3C-SiC or β-SiC) is of particular importance owing to its high mobility (1000 cm^2/Vs), electron saturation drift velocity (2.2×10^7 cm/s), high resistance to oxidation, high thermal conductivity and low expansion coefficient. It is also among the most suitable substrate materials for GaN heteroepitaxy [6,7]. Because of the facts that β-SiC wafers are very costly and limited in availability, and to combine the benefits of the extraordinary physical properties of SiC with the well-developed silicon-based technologies, it is necessary to explore heteroepitaxial growth of β-SiC on Si substrates. A variety of methods, mainly molecular beam epitaxy [8–10], chemical vapor deposition (CVD) [11–14], sputtering [15–18], and ion implantation [19,20] have been employed to obtain thin layers of β-SiC on Si. Films produced at low temperatures are usually amorphous and the formation of good quality SiC films usually requires high temperatures as in case of CVD. PLD, which has long been used for very promising growth of high temperature superconductors, is potentially capable of producing thin layers of a variety of semiconductors. PLD offers the advantages of stoichiometric transfer between target and deposited films, high deposition rates, and growth of well adhered epitaxial layers on different substrates. As far as PLD of SiC is concerned, it is shown to result in crystalline, polycrystalline or amorphous films at relatively low temperatures [21–36]. During the early stage of development of PLD to deposit SiC, nearly stoichiometric polycrystalline layers of β-SiC were grown on Si substrates at 800 °C by Balooch et al. [21]. Since then a number of researchers have

* Corresponding author. Tel.: +91 11 26893955; fax: +91 11 26893666.
E-mail address: yashpalk@gmail.com (Y.S. Katharria).

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reported on SiC films deposition under various substrate, ambiance and post-annealing conditions. Rimai et al. [22] showed the formation of crystalline β-SiC layers for a deposition temperatures above 800 °C. Zehnder et al. [23] deposited epitaxial β-SiC films on Si(100) substrates at a minimum temperature of 900 °C. Soto et al. [24] did an ambiance dependent study of SiC films growth and found that SiC films deposited under Ar environment had better stoichiometry than those grown under nitrogen or vacuum conditions. Polycrystalline films are shown to develop at 700 °C on Si and at 750 °C on sapphire substrates [25]. Huang et al. [26] showed the transformation of PLD grown amorphous SiC films into crystalline during vacuum annealing. Recently, Chauhan et al. [27] demonstrated the application of PLD to grow well adhered SiC films with a good coverage on a heat resistance material.

Most of the early reports on PLD of SiC films have employed a substrate temperature near 800 °C and usually a large density of defects such as stacking faults exists at SiC/Si interface due to a difference of ~8% between the thermal expansion coefficients of the two materials. Therefore, studies utilizing comparatively lower substrate temperatures could be more easy and useful. In the present study, we report a systematic study of fabrication of SiC films on Si(100) substrates by PLD at medium substrate temperatures where most of the substrates, even those with low sublimation or melting temperature could be used. Post-annealing effects under inert environment have also been investigated on these films.

2. Experimental

A KrF excimer laser ($\lambda = 248$ nm, pulse width = 20 ns) was used to ablate SiC target with 99.9% purity (procured from SCI-Engineered Materials, USA). Energy density of the laser pulses was approximately 5 J cm^{-2} with a laser spot of 2 mm × 1 mm on the target. The target-to-substrate separation was 30 mm and the laser pulse frequency was 10 Hz. During ablation, laser light was made to fall on the target at an angle of 50° with respect to the surface normal while Si(100) substrates were kept along the target normal. Prior to deposition, the chamber was evacuated to low 10^{-6} Torr vacuum level and the pressure increased up to 5×10^{-6} Torr during deposition. Before deposition, Si substrates were degreased using trichloroethylene, acetone and methanol and were passivated in 5% HF solution. A substrate temperature of 370 °C was maintained during deposition to achieve an initial short-range ordering of SiC in the as grown films and to get good adhesion. Substrate temperature was monitored using a K-type thermocouple attached to the substrate holder between two Si pieces. Morphology of the as deposited films was investigated by atomic force microscopy (AFM) using a NanoScope IIIa system from Digital Instruments. X-ray photoelectron measurements were performed using a photoelectron spectrometer equipped with an Omicron electron analyzer model EA 125D to study stoichiometry and atomic bonding in the as-grown films. Both the survey scans and core level spectra were recorded using Al Kα radiation. A film grown uniformly on a 1 cm × 1 cm Si piece was cut into four equal pieces. These pieces were then isochronally annealed at 800 °C, 1000 °C and 1200 °C under flowing Ar gas for 2 h keeping one piece as a reference for other studies. Temperature was raised at a rate of 4°/min during annealing. Fourier transform infrared (FTIR) and micro-Raman measurements were also performed on the films using a NEXUS 670 FT-IR system working in 225–4000 cm^{-1} region and a Renishaw Raman spectrometer working with an excitation wavelength of 514 nm from Ar ion laser. Glancing angle X-ray diffraction (GAXRD) was carried out to study the crystallinity development in the films as a function of annealing temperature.

3. Results

3.1. X-ray photo-electron spectroscopy (XPS)

Thin film growth by PLD is demonstrated to result in stoichiometric layers because of ablation of target material by the huge amount of energy deposited by short-duration laser pulses. However, grown SiC films usually contain graphitic (sp^2) and diamond (sp^3) type carbon (C) clusters [21,32,33]. To study the composition of the as-deposited film and look for the possibility of formation of C-clusters, XPS measurements were performed and the results are shown in Fig. 1. Spectra were obtained after an initial 5 min of sputtering of the film using 2 keV Ar^+ ions, which could have removed a layer of around 10 Å, to dislodge surface contaminants. Spectrum of C 1s core line is characterized mainly by two Gaussian peaks centered at 283.2 eV and 285.1 eV. These peaks corresponds to C bonded as Si–C and C-clusters (sp^2 and sp^3), respectively. A weak peak at 287.1 eV is also observed and could have its origin in C bonded with oxygen. Binding energy of Si 2p electrons (100.2 eV), which is shifted by 1 eV from the core 2p level of bulk Si, also confirms the formation of Si–C bonds in the films. A peak near 102.5 eV indicates the formation of Si–O bonding in the film. Detailed analysis by taking into account the integrated intensities of Si and C lines and their atomic sensitivity factors shows that the films contain a 1.7 times larger C fraction as compared to that of Si. However, the ratio of C and Si fractions bonded as SiC is around 1.1, indicating the formation of almost stoichiometric Si–C precipitates driven by the impingement of the energetic particles onto the substrate surface.

3.2. X-ray diffraction (XRD)

Fig. 2 shows the results of XRD measurements performed on annealed SiC films. XRD pattern of the film annealed at 1000 °C is characterized by a broad XRD peak at $2\theta = 35.61°$. This peak corresponds to X-rays diffracted from (111) planes of β-SiC (lattice spacing = 2.5193 Å, lattice constant $a = 4.52$ Å) and supposed to be the strongest