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Structural, electrical and optical properties of nanocrystalline silicon thin films deposited by pulsed laser ablation

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ABSTRACT

Nanocrystalline thin films of silicon (Si) were grown on glass substrates using pulsed laser deposition technique. Si nanoparticles thin films were investigated employing X-ray diffraction, field emission scanning electron microscopy, atomic force microscopy, low temperature electrical transport measurements, and UV–visible absorption spectroscopy. XRD data show that Si films are polycrystalline having cubic crystal structure with preferred (111) orientation. SEM and AFM micrographs exhibit dense and slightly rough surface morphology with well defined columnar grains. Temperature dependence of the resistivity has been fitted with $\ln \rho(T) \propto T^{-1/4}$ which indicates three dimensional variable range hopping conduction mechanism as well as semiconducting behavior of films. Finally, the optical absorbance edge of Si thin films is described in terms of indirect transition model proposed by Tauc in the strong absorption region, and according to Urbach's rule in the medium absorption region.

1. Introduction

Recently, nano-sized materials have attracted great attention because of their unique structure and modified optical, electrical, electronic and magnetic properties compared to their bulk counterparts [1,2], making them key structural blocks for a new generation of electronic, sensor and photonic materials and devices. The unique properties of nanoparticles are strongly dependent on their size, morphology and structure. Silicon (Si) nanocrystals at room temperature [3] have attracted much attention within the scientific community due to their potential applications in silicon based light emitting devices. However, when silicon is in the form of low dimensional structure such as porous silicon, silicon superlattice, silicon quantum dot or nanocrystal, its electrical and optical properties are greatly modified to that of bulk silicon. One of the most interesting effects of low-dimensional semiconductor structures is their size-dependent energy band gap [4–8].

There have been many studies with Si nanoparticles including nano floating gate memory (NFGM) [9] and single electron transistor (SET) [10] for electronic devices, and secondary batteries [11,12], super capacitors and solar cells for energy conversion devices. Silicon thin films can be grown by various deposition techniques, including sputtering, pulsed laser deposition (PLD), chemical vapor deposition (CVD), and molecular beam epitaxy (MBE). Among them, pulsed laser deposition (PLD) technique is found to be an excellent choice due to its simplicity and advantages [13]. In fact, it allows low temperature deposition due to the high energy content of ejected species. Moreover, it has the ability to congruently transfer the stoichiometry from the target to the film, and allows the growth of complex materials. Although a lot of research work is reported on silicon nanostructures, but the investigations towards the complete understanding of various mechanisms governing the physical properties of silicon nanostructures are still under way. In the present work, we applied PLD technique to deposit nanocrystalline silicon films

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on glass substrates and extensively investigated them with respect to their structural, electrical and optical properties.

2. Experimental

Silicon thin films were grown on glass substrates at 5 mTorr Argon background pressure and 650 °C substrate temperature using PLD technique. Before deposition, glass substrates were treated with acetone and ethanol, and then dried in nitrogen gas flow. The base pressure of chamber was kept $\sim 2\times 10^{-8}$ Torr. During deposition, KrF excimer laser (COMPex Pro 205, $\lambda=248$ nm, pulse width=20 ns), operated at 45 mJ laser beam energy was used to ablate commercially available single crystal unpolished silicon wafers (purchased from MTI Corp.). The laser beam, passing through a UV grade quartz window was focused at the target with an incidence angle of 45°. The ablated species of silicon were ejected with high kinetic energies and deposited on substrates rotating in off-axis position with respect to silicon plume normal. Argon gas (99.99% purity) was purged into the chamber through a mass flow controller and variable leak valve to a fixed pressure. After deposition, the samples were cooled to room temperature at a rate of ~5 °C/min. A thickness monitor (Edward FTM7) was used to control and measure the thickness of films at about 500 nm.

Phase purity and crystal structure of the films were investigated by means of X-ray diffraction (PANalytical X'Pert) using CuKα radiation with an accelerating voltage of 45 kV and a current of 40 mA. The surface morphology of the films was characterized by scanning electron microscopy (JEOL SM-5610LV), and atomic force microscopy (Vecco Multimode, Nanoscope V). Current–voltage (*I–V*) measurements were made in a specially designed sample holder where nitrogen gas was used to maintain desired low temperature, and a vacuum of 10^{-3} Torr was maintained. For electrical resistivity measurements, the planer geometry of films (length ~1.5 cm, electrode gap ~5 mm and thickness ~500 nm) was used, and indium contacts were deposited on the films to act as electrodes. Optical parameters were determined from absorption data recorded by UV–visible spectrophotometer (Shimadzu UV-2100) at normal incidence in the range of 190–900 nm.

3. Results and discussion

X-ray diffraction pattern of silicon nanostructured thin film is presented in Fig. 1. The XRD spectra of as-deposited Si films reveal that high quality polycrystalline Si films, with cubic crystal structure and preferred (111) orientation have been obtained. The crystallite size of thin film was calculated using Debye–Scherrer equation [14]

$$D=\frac{0.9\lambda}{\beta\ \cos\ \theta} \tag{1}$$

where D is the diameter of crystallite in the film, λ is the wavelength of incident X-rays, β is the full width at half maximum (FWHM) in radians, and θ is the Bragg's angle. The crystallite size of Si thin films was estimated to be 9.2 nm. The prominent peak at $2\theta\approx\approx 28^{\circ}$ is assigned to (111) orientation in cubic silicon. The lattice constant (a) of the film material (Si) is found to be 5.421 Å. Ayouchi et al. [15] prepared silicon thin films by PLD at different deposition temperatures (400–800 °C) and reported the value of lattice constant between 5.46 and 5.53 Å. In order to study the evolution of microstrain and crystallite size, we used the Williamson–Hall equation [16]

$$\beta\ \cos\ \theta=\frac{0.9\lambda}{D}+2\varepsilon\ \sin\ \theta \tag{2}$$

where β is the full width at half maximum of XRD peaks, D is the crystallite size, λ is the X-ray wavelength, θ is the Bragg diffraction angle and ε is the micro strain present in the lattice. In this case, the term $\beta\ \cos\ \theta$ is plotted with respect to $\sin\ \theta$ for all diffraction peaks of silicon nanoparticles as shown in Fig. 2. On drawing a linear extrapolation to this plot, the intercept gives the crystallite size, while the slope gives the micro strain. The average crystallite size and the micro strain of the sample have been found to be 9.08 nm and 1.97×10^{-3}, respectively.

Fig. 3 presents the SEM image of silicon nanostructured thin film. The morphology of sample looks like columns. It is clear from micrograph that the surface is dense and slightly rough. The uniformity of nanocrystal size distribution has

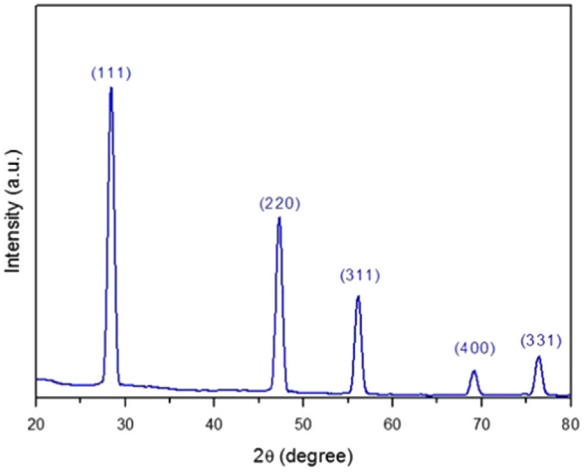

Fig. 1. XRD spectra of Si nanostructured thin film.

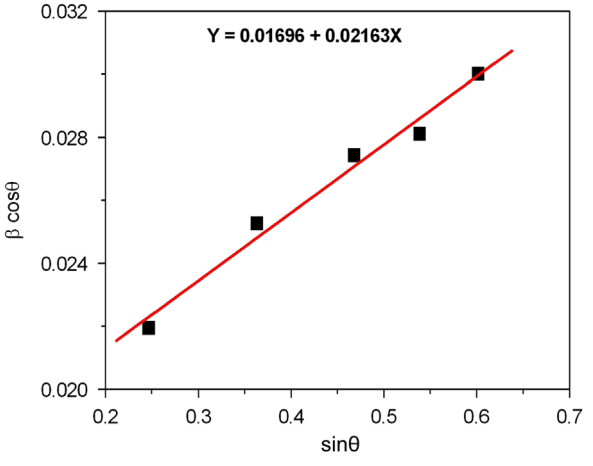

Fig. 2. Williamson–Hall plot of Si nanostructured thin film.