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Si nanostructures grown by picosecond high repetition rate pulsed laser deposition

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ABSTRACT

One-step growth of n-doped Si nanostructures by picosecond ultra fast pulsed laser deposition at 1064 nm is reported for the first time. The structure and morphology of the Si nanostructures were characterized by X-ray diffraction, scanning electron microscopy and atomic force microscopy. Transmission electron microscopy studies revealed that the shape of the Si nanostructures depends on the ambient argon pressure. Fibrous networks, cauliflower formations and Si rectangular crystals grew when argon pressure of 300 Pa, 30 Pa and vacuum (10^{-3} Pa) conditions were used, respectively. In addition, the electrical resistance of the vacuum made material was investigated.

1. Introduction

In the recent years, Si nanostructures have attracted significant attention due to their potential application in microelectronics, optoelectronics [1,2] and medicine [3]. Si nanostructures are used in a wide range of applications that varies from third-order nonlinear optical response when dispersed in organic solvent [4] to lithium battery storage [5–7]. However Si due to its indirect energy band gap has low luminescence which is attributed to its long radiative lifetime [8,9]. For applications such as quantum computing [10], light emitting diodes (LEDs) [8,11] and lasers [8], small nanoparticle size and low electron density are required in order to increase quantum confinement of Si. Si thin films [12,13], crystals [14,15], nanoparticles [16–20], nanowires [21,22], nanoclusters [23] and even nanorings [2] have been prepared in the past using advanced laser techniques with the conventional pulsed laser deposition (PLD) method to be the most popular. Unlike, conventional PLD, where long pulses, typically >5 ns, of few hundred mJ and low laser repetition rates are being used, we employed ps high repetition rate PLD also known as ultra fast PLD (UFPLD) to produce Si nanostructures in different ambient Ar pressures.

2. Experimental

Glass and n-type (1 0 0) Si substrates were cleaned ultrasonically in a sequence of acetone, methanol and isopropanol and blown dried with high purity N_2 gas. The substrates were used for the deposition of Si nanostructures at room temperature and under three different ambient pressures: (i) 300 Pa and (ii) 30 Pa of argon pressure (P_{Ar}), and (iii) vacuum of 10^{-3} Pa. The PLD chamber was initially evacuated to a base pressure of 10^{-4} Pa, with the aid of a turbomolecular pump, prior to introducing the high purity argon gas. The fundamental wavelength, 1064 nm, from a Nd:Vanadate ($Nd:YVO_4$) laser was used for the ablation of a n-type (1 0 0) Si wafer target. About 240×10^6 pulses of 10 ps pulse width at full width half maximum and repetition rate of 200 kHz irradiated the Si target. The laser irradiance was kept constant at 7.71×10^9 W cm^{-2} and the target was rotated and rastered continuously during UFPLD. The Si target-to-substrate distance remained at 1.5 cm for all depositions.

The as-grown Si nanostructures on glass substrates were structurally and morphologically characterized by X-ray diffraction (XRD) analysis, SHIMADZU, XRD-6000 and atomic force microscopy (AFM), Ambios tech., Q-Scope Series Scanning Probe Microscope, respectively. The topography of the samples was imaged by SEM, Tescan Vega LSU. Transmission electron microscopy (TEM) in dynamic contrast imaging mode was used to investigate the morphology and structure of the Si nanostructures at the atomic scale. A Jeol 2011 electron microscope operated at 200 kV with a point resolution of 0.194 nm and spherical aberration coefficient C_s = 0.5 mm was employed. The resistance of the vacuum made Si

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nanostructures was measured using a four probe technique, over the temperature range 200–300 K. Both positive and negative biases were used for the measurements. The electrical measurements were performed in a Quantum Design PPMS system.

3. Results and discussion

Crystalline Si nanostructures were grown on glass substrates using both Ar ambient and under vacuum conditions. In the XRD patterns of Fig. 1, we can observe the reflections (1 1 1) and (2 2 0) of Si from the samples produced at P_{Ar} of 30 Pa and 300 Pa, indicating that crystalline structures were obtained at room temperature. Even though, the Si sample fabricated at 10^{-3} Pa seems to be amorphous based on the XRD pattern of Fig. 1, TEM microscopy revealed that the sample is partially crystalline, the crystallites are shown in Fig. 3c. No XRD reflections were observed from the Si nanostructures produced under 10^{-3} Pa.

Fibrous Si web-like formations were produced on n-type Si substrates at P_{Ar} = 300 Pa, Fig. 2a. Upon decrease of the Ar pressure to P_{Ar} = 30 Pa, cauliflower Si nanostructures were grown, seen in Fig. 2b. When vacuum conditions (10^{-3} Pa) were used during the UFPLD process rough Si nanostructures were obtained, Fig. 2c. The inset in Fig. 2c shows an AFM image of the Si nanocrystals grown under vacuum conditions with RMS roughness of 42 nm.

TEM revealed that the shape and size of the Si nanostructures produced under different Ar pressures vary significantly. Spherical

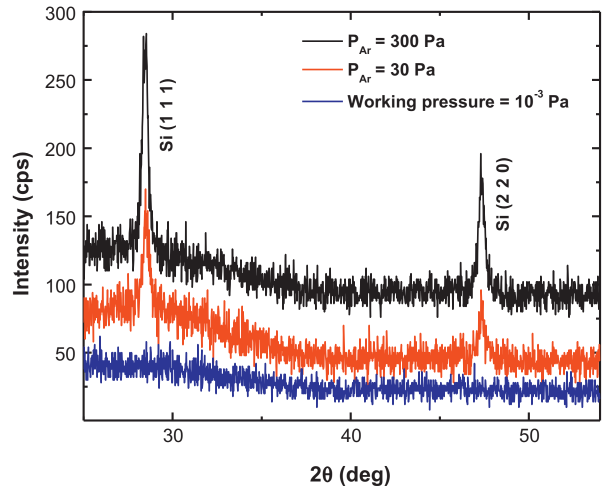

Fig. 1. XRD patterns from Si nanostructures produced on glass substrates using P_{Ar} = 300 Pa, P_{Ar} = 30 Pa and vacuum (10^{-3} Pa) conditions.

crystalline Si nanoparticles embedded in an amorphous Si matrix were produced for the ambient of 300 Pa, seen in Fig. 3a. The spherical morphology and the different size of the Si nanocrystals are evident. These spherical Si nanostructures are forming the fibrous networks shown in Fig. 2a. The reduction of P_{Ar} resulted in the

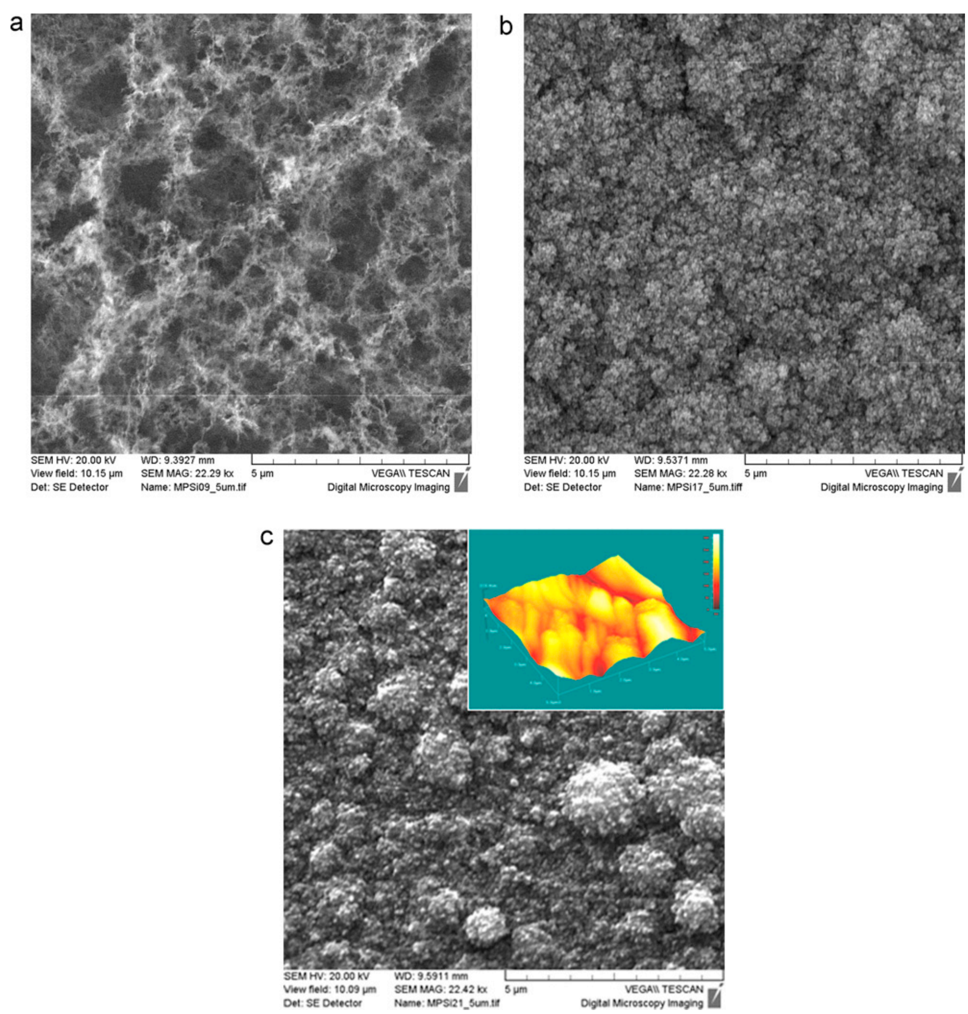

Fig. 2. SEM images from Si nanostructures synthesized on n-type Si substrates at (a) P_{Ar} = 300 Pa (fibrous web-like formations), (b) P_{Ar} = 30 Pa (cauliflower structures) and (c) under vacuum conditions (rectangular crystals). Inset in (c): AFM image of Si nanocrystals grown under vacuum conditions.