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Ion-beam-induced nanodots formation from Au/Si thin films on quartz surface

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ABSTRACT

We report the synthesis of Si nanodots on quartz surface using ion irradiation. When a bi-layer of ultra-thin Au and Si on quartz surface is irradiated by 500 keV Xe-ion beam, the bi-layer spontaneously transforms into nanodots at a fluence of 5×10^{14} ions cm^{-2}. The spatial density and diameter of the nanodots are reduced with increase in applied ion fluence. The nanostructures exhibit photoluminescence in the visible range at room temperature where the intensity and wavelength depends upon ion fluence. The observed evolution seems to be correlated to ion beam mixing induced silicide formation at Au–Si interface.

1. Introduction

Si nanostructures have attracted wide interest for long time, owing to their novel properties like size dependent photoluminescence, electrical properties, possibility of surface functionalization etc. Si nanoparticles are promising for application in light emission, in photovoltaics and electronics, photocatalysis, etc., which are summarized in a recent review by Huan and Shu-Quing [1]. Optical properties of Si nanostructures, for instance, are addressed for long time since the indirect nature of the band gap of Si hinders optical emission, yet the compatibility of Si in optoelectronic devices demands enhancement of emission efficiency. Since the observation of visible range photoluminescence [2] from nanoporous Si, different techniques are undertaken to fabricate Si nanostructure [1]. Moreover, synthesis of an array of Si nanostructure on the surface of an insulator has received particular attention in the context of device applications [3–5]. Dewetting of Si layer on SiO_2 during high temperature annealing is shown to result in formation of Si islands on SiO_2 with submicrometer dimension [3]. Annealing of substoichiometric SiO_x ($x < 1$) produced Si nanocrystals in silica, where the nucleation and growth of nanocrystals depends upon degree of supersaturation of Si in SiO_2 [4]. A two dimensional array of Si nanodisks on SiO_2 was fabricated by Huang et al. [5]. Their method, however, comprised of several chemical steps, etching, and subsequent lift-off. On the other hand, lithographic process to design particular array on a polymer film and subsequent deposition of Si is limited in terms of large scale synthesis. Thus, in the context of synthesis of Si nanostructure array on insulator, a single-step synthesis with a control over dimensions and areal densities of nanostructures is still a very important objective under investigation. In this direction, ion beam synthesis can be a highly effective route because it offers desirable advantages such as single step synthesis over large area, controllability of nanostructures through a number of independently variable experimental parameters, such as ion energy, fluence, etc. In fact, ion implantation has been successfully used for synthesis of a two-dimensional array of Si nanoparticles embedded in insulators like SiO_2 [6,7], which involves an annealing step subsequent to implantation or use of a stencil mask. Moreover, nanodot structures were seen to evolve in case of low energy (few hundreds of eV) ion irradiation on metal coated Si substrates [8–10], which was assumed to be driven by formation of metal silicide at the metal/Si interface, indicating a route for ion beam synthesis of Si nanostructure mediated by a metal top layer. This is further important in relation to nanotechnology for metal rich Si nanostructure synthesis [11].

In this study we present a synthesis technique of Si nanodot structure on quartz surface using irradiation of energetic ions. A bilayer of Au and Si is deposited on quartz surface. A nonodot array forms spontaneously on the quartz surface when 500 keV Xe-ions are irradiated on the metal–semiconductor bilayer. Present experiment also indicates a tunability of nanodot morphology through

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ion fluence. In addition, a fluence dependent enhancement of photoluminescence of the irradiated samples is also evident. Thus, our experimental study indicates a way for self-organized nanodot synthesis in a single step.

2. Experimental

The depositions of Si and Au thin films were carried out using e-beam evaporation technique in a high vacuum deposition unit (MANTIS) having a base vacuum of 2×10^{-7} mbar. First a 6 nm Si film and then a 6 nm Au film was deposited on high quality quartz substrates. The samples were irradiated by 500 keV Xe-ions at the Low Energy Ion Beam Facility (LEIBF), IUAC, New Delhi. The ion flux was kept constant around 1 μm cm^{-2} and the beam was scanned over the samples in a raster pattern to achieve homogeneous irradiation. The morphology of as-deposited and ion-irradiated samples was investigated using atomic force microscopy (AFM) in tapping mode. AFM images were analyzed using WsXM [12] and Gwyddion software [13]. Corresponding compositional variation was studied using energy dispersive X-ray spectroscopy (EDS) (Oxford Instruments), carried out using 20 keV electron beam in a scanning electron microscope (SEM) (Carl Zeiss). The photoluminescence (PL) spectra are obtained at room temperature (RT) in a steady state fluorescence spectrometer (Edinburgh Instruments) using the 325 nm line of a He–Cd laser as primary excitation.

3. Results and discussion

Representative AFM images of the as-deposited and Xe-ion irradiated bi-layers of Au–Si thin films are shown in Fig. 1(a–c). The as-deposited bi-layer shows a granular structure, where few bigger grains, having lateral dimension greater than 150 nm, can also be seen. In fact, formation of a granular structure can be expected in the present case because of the thin Au layer deposition on top of Si. For such small thickness of the deposited films, deposited Au form interconnected islands on the surface. However, a dramatic modification of the surface morphology takes place subsequent to 500 keV Xe-ion ion irradiation. At the lowest fluence of 5×10^{14} ions cm^{-2}, the AFM micrograph reveals formation of nanodots on SiO_2 surface. We do not see any specific spatial order or pattern of arrangements of the dots on the surface. Further development of the nanodot morphology as a function of applied fluence can be followed from the AFM image shown in Fig. 1(c), corresponding to the fluence of 1×10^{16} ions cm^{-2}. Indeed, the nanodot morphology observed in Fig. 1(c) is reduced to a few nanodots scattered over the surface. It can be inferred that it is a result of sputtering of nanodots at this higher fluence. The morphological evolution of the irradiated samples is also reflected in the rms roughness determined from the AFM images and reported in Fig. 1(d). The rms roughness increases with the formation of nanodots and subsequently decreases to a much lower value of 0.72 nm when the nanodot morphology is sputtered out at the highest fluence of 1×10^{16} ions cm^{-2}.

The diameter distribution of the ion-beam-induced nanodots is determined by using ImageJ software [14]. The distribution of the nanodot diameters is depicted in Fig. 1(e) and (f), for ion fluences of 5×10^{14} and 1×10^{16} ions cm^{-2}, respectively. It is seen from Fig. 1(e) that the particle diameters cover a range from around 23 nm to 150 nm, whereas for higher fluence it reduces to a range of 20 nm to 65 nm as shown in Fig. 1(f). The red dashed line in Fig. 1(e) and (f) is a Gaussian fit to the experimentally obtained diameter-distribution. It is observed that the peak of the distribution is at 70 nm, corresponding to an average diameter of nanodots. At the highest fluence, the dot diameters are reduced to 45 nm, as is evident from Fig. 1(f).

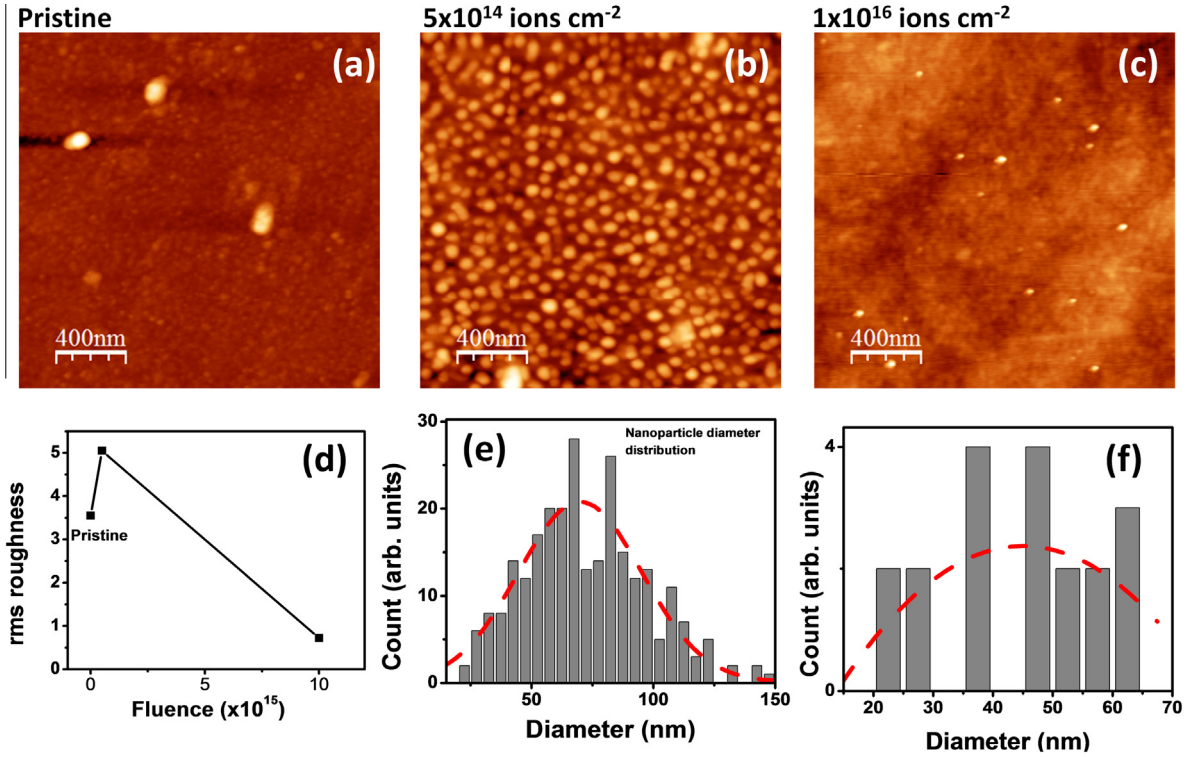

Fig. 1. AFM images of the (a) pristine sample and samples irradiated to the fluences of (b) 5×10^{14}, and (c) 1×10^{16} ions cm^{-2}, (d) rms roughness of the pristine and irradiated samples, (e) and (f) are diameter distributions of nanodots observed at the fluences of 5×10^{14} and 1×10^{16} ions cm^{-2}, respectively. The dashed red lines in (e) and (f) represent the Gaussian fits to the data.