Vacuum 86 (2011) 96–100

Contents lists available at ScienceDirect

Vacuum

journal homepage: www.elsevier.com/locate/vacuum

Nano/micro-structuring of oxide thin film under SHI irradiation

R.S. Chauhan [a,*], D.C. Agarwal [a,b], S. Kumar [c], S.A. Khan [b], D. Kabiraj [b], I. Sulania [b], D.K. Avasthi [b], W. Bolse [d]

[a] Department of Physics, R. B. S. College, Agra 282 002, India
[b] Inter-University Accelerator Centre, New Delhi 110 067, India
[c] Department of ASH (Physics), FET, Manav Rachna International University, Faridabad 121 001, India
[d] Institut fuer Strahlenphysik, Universitaet Stuttgart, Allmandring 3, 70569 Stuttgart, Germany

A R T I C L E I N F O

Article history:
Received 6 July 2010
Received in revised form
14 February 2011
Accepted 26 April 2011

Keywords:
Nano patterning
Oxide thin film
Ion irradiation
Atomic force microscopy

A B S T R A C T

In the present work, we have studied the nano/micro-patterning of the surface of NiO thin films on different substrates (SiO_2, Si and Al) using 100 MeV Ag ions at LN_2 temperature and at an incidence angle of 75° with the beam axis. The surface morphology of the irradiated surface is observed by Atomic force microscopy (AFM). AFM images of ion beam irradiated samples show the restructuring of initially flat and coherent NiO film into an almost periodic NiO lamellae structure. The quite regular lamellae with width, height and average distance of hundreds of nm are oriented perpendicular to the beam direction. Section analysis of the AFM images reveal that the width of the lamellae is less in case of NiO films deposited on SiO_2 substrate in comparison to Al substrate. The cracking and the development of lamellae structure is observed at higher fluence in the case of Al substrate in comparison to other substrates.

1. Introduction

The structuring and reorganization of the surface by ion sputtering has been recognized from earlier time [1]. The fabrication of periodic surface structures varying from nm to mm with a spatial periodicity has attracted considerable attention to the current research and technology because of its potential as templates for the fabrication of nanostructured materials [2]. The swift heavy ion irradiation is an efficient technique to manufacture the sub-micron to nanoscale surface structures. In the low energy regime, sputtering leads to the erosion of surface by elastic collision of ions with atoms in the solid during ion bombardment, which results in the formation of nanoscale pattern on the surface [3–5]. Swift heavy ion (SHI) can modify the properties of the materials and it has been shown that SHI anneal the defects produced by the low energy ions [6,7]. Moreover, there are few reports, which reveal that the semiconductor surface can be amorphized and roughened in a highly localized region around the ion path, which results in different scenario of the surface under SHI irradiation [8]. Recently, it has been shown that self-organization of thin film surfaces during high-energy heavy ion irradiation results in structure formation at a sub-micrometer scale, creating peculiar surface morphologies of quite regular patterns. Bolse et al. showed the self-organization of surface of thin oxide film under SHI irradiation [9,10]. They have also shown the dewetting of oxide thin film on Si [11] under ion irradiation, which is similar to the dewetting pattern of liquid thin film [12,13]. In the low fluence regime, the damaged zone produced by SHI is localized within a diameter of a few nm. In most of the cases one-to-one correspondence is observed in the number of ion impact and the surface damage. The surface features observed after irradiation are craters, structural disorder and surface point defects [14]. In some cases the surface damage is observed due to the cumulative effect of ion irradiation, which means that two or three ions incident within the same spatial region and produce a track [15]. In the high fluence regime, damaged zone with dimensions of a few micrometers was expected as a result of overlapping of the damaged zones created by the single ion. When a swift heavy ion strikes on the surface of a solid, it slows down via two processes (i) direct transfer of recoil energy to target atoms through elastic collisions i. e. nuclear energy loss (S_n) and (ii) electronic excitation and ionization of target atoms or inelastic collisions i. e. electronic energy loss (S_e). The later process is dominant in the case of SHI. If the thickness of the targeted medium is sufficiently smaller than the range of the projectile ion, the energy deposition is mainly due to the electronic energy loss, which is also called the linear energy transfer (LET). This energy deposition leads to production of defects and self-organization of the surface leading to change in surface roughness and nanoscale structures in some conditions.

In the present study, NiO thin films, deposited on different substrates, were irradiated by 100 MeV Ag ions at LN_2 temperature and at an incidence angle of 75° with respect to the beam axis. We

* Corresponding author.
E-mail address: rvschauhan@yahoo.com (R.S. Chauhan).

0042-207X/$ – see front matter
doi:10.1016/j.vacuum.2011.04.024

observed that after the application of high fluence, the surface of the NiO films is re-organized in the form of periodic lamellae structures.

2. Experimental

NiO thin films were deposited on Si, Al/Si and SiO_2/Si by rf-magnetron sputtering at Stuttgart University, Germany. The Al film of thickness 250 nm on Si was deposited by resistive heating method. The SiO_2 layer was prepared by annealing the Si substrates in presence of oxygen at Inter-University Accelerator Centre (IUAC), New Delhi. The irradiation has been performed at LN_2 temperature and at an incidence angle of 75° with respect to beam axis using the 100 MeV Ag ions from 15 UD Pelletron accelerator at IUAC, New Delhi. The samples were mounted on the irradiation ladder in high vacuum irradiation chamber. In order to do homogeneous irradiation, the ion beam was carefully scanned over an area (5 mm × 5 mm). The films were irradiated at different fluences 1×10^{13}, 3×10^{13}, 7×10^{13}, 1×10^{14}, 3×10^{14}, and 6×10^{14} ions/cm^2. The electronic stopping power of 100 MeV Ag ions in NiO thin film is 20.17 keV/nm while nuclear stopping power is 0.12 keV/nm. Since the range of the 100 MeV Ag ions (9.22 μm) was much greater than the film thickness, no ions were implanted in to the film and modification was expected only due to the S_e.

Surface morphology of irradiated and pristine samples was studied by Atomic force microscopy (AFM). The sputtering of Ni and O from the films is determined by on-line elastic recoil detection analysis (ERDA) measurement. The sample was kept at LN_2 temperature and tilted at 75° with respect to the beam axis for ERDA measurement. The recoils were analyzed by large area position sensitive detector telescope (LAPSDT) placed at a scattering angle of 45°.

3. Results and discussion

The irradiation at small angle of incidence with surface or large angle of incidence with beam axis results in restructuring of crystalline NiO films. The AFM study shows that the continuous film of NiO starts to crack at low fluence perpendicular to the beam direction and after the application of high fluence the material between the cracks begins to shrink and self-organize into the periodic lamella structures. Fig. 1(a)–(c) show the AFM micrograph of the NiO thin film on different substrates irradiated at fluence of 6×10^{14} ion/cm^2. The width of the lamellae is less in the case of NiO thin film deposited on SiO_2 substrate and greater in the case of Al substrate. The width and separation of lamellae are given in Table 1. It is also observed that the cracking and development of lamellae structure occurs at higher fluence in the case of Al substrate. The development of lamellae structure with fluence for SiO_2 and Al substrates are shown in Fig. 2 and Fig. 3 respectively. The possible mechanism of self-organization of NiO thin film can be understood by visco-elastic model followed by thermal spike model. When swift heavy ion passes through the material, it looses energy via electronic excitation or ionization and produces a cylindrical zone of few nm. The electronic subsystem comes in to equilibrium by electron–electron collision and electron-phonon coupling and produces the Gaussian like temperature profile in the vicinity of ion path in the material [16–18]. High temperature causes the transient local melting of the lattice within a cylindrical zone. The energy rapidly dissipates into the cold surroundings resulting in the solidification. The as-deposited films of NiO were polycrystalline in nature and it is expected that film is amorphized after the high fluence of 1×10^{13} ions/cm^2 at low temperature. In our work of the irradiation of NiO thin film at room temperature shows disordering at high fluence [19]. The melting and subsequent re-solidification of

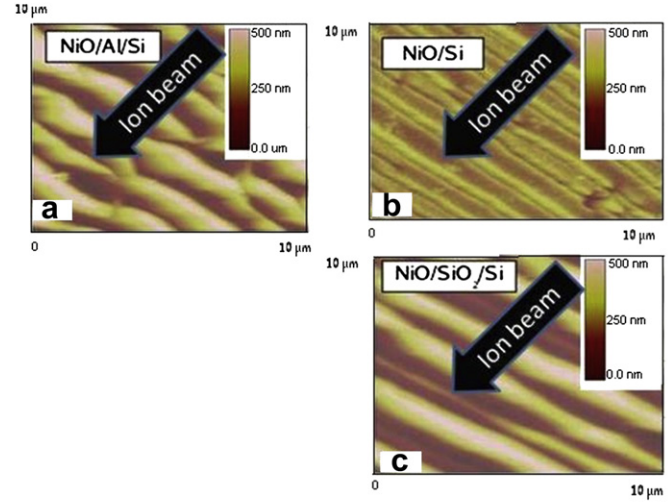

Fig. 1. AFM images of NiO film on different substrate irradiated at 6×10^{14} ions/cm^2 (a) on Al/Si (b) on Si (c) on SiO_2/Si.

materials generates the uniaxial tensile stresses along the ion track [20,21]. When the tensile stresses along the beam direction overcome the fracture strength of the NiO surface, it shows periodic cracking perpendicular to the beam direction due to Grinfeld instability [22,23]. Using linear surface stability analysis, Grinfeld showed that the competition between elastic strain energy and surface energy leads to the amplification of perturbation of specific periodicity on such stressed surface. The formation of cracks release elastic stress energy and consumes energy for the creation of the cracked surface. For each stressed surface there is an optimum cracking distance, which minimizes the total free energy. The theory of lamellae formation can be explained by hammering effect, which was given by Klaumuenzer [24–26]. When fluence increases, the material between cracks shrinks and develops into the lamellae structure. The width of the lamellae decreases and the separation between them increases with the increase of fluence. The possible reason for higher width and lack of periodicity of lamellae in case of Al substrate can be understood on the basis of the energy transferred from the film to the substrate through interface. Since the resistivity of the Al is less than Si and SiO_2, the energy deposited by SHI in the film can be easily transferred to Al substrate.

Since the surface structures induced by ion bombardment are formed due to the interplay between sputtering and surface diffusion, we measure the sputtering of NiO thin film under the swift heavy ion bombardment. The sputtering yield and stoichiometry of the NiO thin film under 100 MeV Ag ions bombardment are determined using the on-line ERDA technique. Areal concentration of O and Ni and the ratio of areal concentration of Ni to O or stoichiometry were calculated from recoil spectra of individual

Table 1

Sample	Fluence (ions/cm^2)	Width of Lamellae	Distance between Lamellae	Height of Lamellae
NiO/Al/Si	1×10^{14}	1.3 μm	450 nm	150 nm
NiO/Al/Si	3×10^{14}	1.0 μm	600 nm	220 nm
NiO/Al/Si	6×10^{14}	0.9 μm	700 nm	310 nm
NiO/SiO_2/Si	3×10^{13}	1.2 μm	300 nm	55 nm
NiO/SiO_2/Si	7×10^{13}	1.1 μm	410 nm	62 nm
NiO/SiO_2/Si	1×10^{14}	1.0 μm	475 nm	175 nm
NiO/SiO_2/Si	3×10^{14}	0.95 μm	605 nm	250 nm
NiO/SiO_2/Si	6×10^{14}	0.80 μm	800 nm	620 nm