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Azimuthal angle dependence of nanoripple formation on Si (100) by low energy ion erosion

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ABSTRACT

Generation of self organized nanoripple patterns on Si (100) single crystal surface using low energy Ar ion beam erosion has been studied. Ion energy and ion fluence dependence of the ripple pattern is in general agreement with the reported works. However, it is found that at relatively low fluences, the pattern formation depends upon the direction of the projection of the ion beam on Si surface with respect to its crystallographic orientation. Ripple formation is facilitated if the projection of ion beam on the sample surface is along (010) direction as against (011) direction. For higher ion fluence, when the Si surface layer is fully amorphized, pattern formation is independent of the azimuthal direction of the ion beam. Copyright © 2013 VBRI press.

Keywords: Nanoripple patterns; ion-beam irradiation; crystallographic orientation; atomic force microscopy.



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Introduction

The fabrication of ordered structures at the nanometer length scale happens to be the basis for many technological applications in a variety of fields. A simple approach for producing nanostructure is the low energy ion beam erosion of solid surfaces. This method has proven to be a flexible self organization method for the generation of well ordered nanostructures on the surfaces of metals, insulators and semiconductors [1-8]. Till now, two different types of self organized patterns during ion beam erosion have been observed, ripples [2, 7] and dot pattern [8, 9] depending on whether ion beam incidence is oblique or normal respectively. It has been suggested that pattern formation at normal incidence of ion beam is assisted by impurity atoms which may get deposited on the surface of the sample intentionally or unintentionally [10-12]. Recently, nanoscale ripple patterns have been used as templates for the deposition of metallic thin films [13-17]. It is shown that, these patterns can induce optical [13-14] and magnetic [15-17] anisotropies in the deposited films.

Systematic studies have been reported in the literature on the evolution of ripple patterns on Si surfaces upon ion irradiation. Variation in wavelength and amplitude of the ripples as a function of ion fluence, ion energy, angle of ion incidence, and for different inert ion species has been studied. In general, for oblique ion incidence, ripple patterns evolve on the surface with wave vector either parallel or perpendicular to the ion beam projection depending on the ion incidence angle [2, 7]. Dot patterns are observed for normal incidence of ions with simultaneous sample rotation [8, 9]. Fine variations in the ion incidence angle can make topographical transition from ripple patterns to other patterns [9]. Wavelength of the ripple in general is found to remain constant with ion fluence, while it increases with ion energy [7, 9]. Ripple amplitude and ordering increases with ion fluence. Mass of the bombarding ions also plays a decisive role in the ripple formation [7]. Extensive studies have also been done on ripple formation on Si surface [5-8]. Despite such detailed studies on the effect of various irradiation parameters on ripple formation on Si surface, no study has been reported in the literature on the dependence of ripple formation on crystallographic orientations of Si substrate with respect to the ion beam direction. Crystallographic orientation is expected to affect the pattern formation at the surface, since various factors like sputtering rate, surface diffusion etc. are expected to exhibit anisotropy with respect to the crystallographic directions. Therefore, in the present work ripple pattern formation on Si (100) surfaces by low energy Ar ion beam erosion has been studied as a function of the azimuthal angle of the projection of ion beam direction on the sample surface.

Experimental

Commercially available polished Si (100) substrates with a root-mean-square (r.m.s) roughness of ~ 0.5 nm have been used directly without any surface treatment for the production of self organized ripple patterns. For ion beam erosion experiments, Kaufman type hot cathode ion source with a beam diameter of 30mm was employed. Base pressure in the deposition chamber was 2×10^{-7} mbar and vacuum during the irradiation was 2×10^{-7} mbar. Ion beam erosion experiments have been carried out with Ar ion at two different energies, namely 500 eV and 1000 eV. The angle of incidence of the ion beam with respect to the surface normal was kept fixed at 65^{0} . The ion current during irradiation was 6mA, which corresponds to an average flux of 2.2×10^{15} ions cm⁻² s⁻¹.

Surface morphology of the sample before and after irradiation was analyzed *ex-situ* by means of Atomic force microscopy using Digital Instruments (Model-Nanoscope-E) in contact mode. XRD- Phi scan has been done using Brucker-D8 diffractometer.

Results and discussion

The in-plane crystallographic orientation of the Si wafer has been analyzed by XRD- Phi scan. For this, scattering angle 2θ was fixed at a value corresponding to the (220) plane of Si. The sample was given a chi tilt of 45° in order to bring the (110) planes normal to the scattering vector. Sample was rotated about an axis perpendicular to the sample surface through 360° . Fig. 1 shows the corresponding XRD-Phi scan. Four peaks are observed corresponding to the (110), (101), (110), (101) planes. This way (010) and (011) direction in the sample plane was identified. Two different samples were irradiated with 500 eV Ar^+ at an oblique angle of 65^0 from the surface normal, and with projection of ion trajectory on the substrate surface being along the (010), or (011) directions. The sputtering was done for different erosion times with an ion flux of 2.2×10^{15} ions cm⁻² s⁻¹ for studying the evolution of ripple patterns along two azimuthal crystallographic axes (010) and (011).



Fig. 1. XRD Phi scan of Si (100) with scattering angle 2θ fixed at a value corresponding to the (220) plane of Si.



Fig. 2. AFM images of Si (100) surface irradiated with different fluences of 500eV Ar ions at an oblique angle of 65^0 from the surface normal. The ion flux was kept at 2.2×10^{15} ions cm⁻² s⁻¹. Figure 2(a) shows the AFM image of an unirradiated Si (100) surface. The projection of ion trajectory on the sample surface was either along (010) azimuthal direction, (b-e), or along (011) azimuthal direction (f-i). Scanned area is 1µm x 1µm in all the cases. Thick arrow shows the direction of the ion beam. The insets show Fourier transform of the images.

Fig. 2(a) shows the AFM image of an unirradiated Si (100) surface, (b), (c), (d) and (e) shows the ion beam irradiated Si (100) samples with ion beam projection along (010), while **Fig.** 2(f), (g) ,(h) and (i) shows the same with ion beam projection along (011) direction, for different ion fluencies. Corresponding Fourier transform of the images have been shown in the inset. With increasing ion fluence ripple patterns with ripple wave vector, k along the projection of the ion beam slowly develops in the both the In addition to the ripple patterns, surface cases. corrugations normal to the k also develops. These corrugations are similar to that observed in several reports [16, 18, 19]. It has been shown that the coarsening of this corrugations follows the same ion fluence dependence that of the ripples. Therefore in the following only the development of ripple pattern is discussed. One may not that for the highest fluence of 1.6×10^{18} ions cm⁻², same quality of the ripple pattern is formed along both (010) and (011) directions. However at lower fluences upto 3.9×10^{17} ions cm⁻², the difference in the pattern formation with (010) and (011) orientation is clearly visible. While at a fluence of 1.3×10^{17} ions cm⁻² the ripple formation is already started in the sample with (010) orientation, in sample with (011) orientation, only a faint orientation of rectangular shaped structures along the direction normal to the projection of ion beam direction is observed. These results clearly show that at low fluences, development of nanopatterns at the Si (100) surface is significantly affected by the direction of projection of ion beam on the sample surface.



Fig. 3. The variations in roughness of the ripple surface and ripple wavelength as a function of ion fluence for both the azimuthal direction; (a) (010) and (b) (011).

Variation in r.m.s roughness, which is a measure of the ripple amplitude, as a function of ion fluence for (010) and (011) azimuthal direction is shown in **Fig. 3**. RMS roughness of the irradiated surfaces was determined using AFM. It is found that roughness of the ripple increases with the ion fluence for both the cases. Wavelength of the ripples is observed to be nearly constant with ion fluence. The ripple wavelength for two different ion energies, 500 eV and 1000 eV was also studied. At 500 eV the wavelength obtained is 35 nm, while for 1000 eV it is 40 nm. These results are in general agreement with earlier reports on the effects of ion energy and fluence on ripple structure [**7**].

The process of ripple formation due to ion-beam erosion is described theoretically by the linear Bradley and Harper (BH) model [20]. According to this model the formation of ripple patterns is due to the instability caused by the competition between curvature dependant sputtering

that roughens the surface and different surface relaxation mechanisms (e.g., surface diffusion). The surface height evolution $h(\mathbf{r}, \mathbf{t})$ can be described by-

$$\partial_t h = v_x \partial_x^2 h + v_y \partial_y^2 h - D \nabla^4 h$$

where v_x , v_y are the effective surface tension coefficients generated by the erosion process and D is the surface relaxation term due to surface diffusion (thermally activated). This equation successfully describes the formation and early evolution of the regular patterns. It is successful in predicting the ripple wavelength and orientation [21]. Further modification in the theory have been done to incorporate some nonlinear effects [22,23] which explain experimental features such as saturation of the ripple amplitude [24,25] the observation of rotated ripples [26], and the appearance of kinetic roughening However, effect of a possible azimuthal [27,28]. anisotropy in surface relaxation processes has not been discussed in any of these theories explicitly. In metallic single crystalline surfaces, a strong dependence of the shape of the patterns on the crystallographic orientation of the substrate have been observed, which may be due to the along different diffusion constants different crystallographic directions [29-31]. However, Si is expected to get amorphized at relatively low ion fluence [32, 33]. Therefore at high fluences at which the ripple structure is well developed, one does not expect any dependence of the ripple patterns on the direction of projection of ion beam on the substrate surface [8,9]. This is in conformity with the experimental observations. However at low fluences when the surface layer of Si (100) is not fully amorphized, some anisotropy in the pattern formation is evident, depending upon whether the projection of direction of ion beam is along (010) or (011) direction. Present work shows that ripple pattern formation is facilitated, if the projection of ion beam is along (010) direction.

Conclusion

Formation of nanorippled structure at the surface of silicon (100) single crystal due to low energy ion beam erosion has been studied. Si (100) surface is irradiated with 500eV Ar ions at a grazing incidence of 65° from the surface normal, with different values of the azimuthal angle of the projection of ion beam direction on the sample surface. At a sufficiently high fluence of 1.6×10^{18} ions. cm⁻², well defined ripple pattern is formed irrespective of the azimuthal direction of the ion beam. However, at low fluences a significant difference can be seen in pattern development depending upon whether the projection of the ion beam on sample surface is along (010) or (011) direction. A regular ripple pattern starts developing at a lower fluence when the projection of the ion beam is along (010) direction. A possible anisotropy of surface diffusivity of atoms in crystalline Si may be a possible cause of this difference. However, once the Si surface layer gets amorphized as a result of ion irradiation, this anisotropy is expected to disappear, leading to ripple formation which is independent of the azimuthal projection of the ion beam.

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Reference

- Valbusa, U; Boragno, C; Buatier de Mongeot, F. J. Phys.: Condens.Matter 2002, 14, 8153.
 DOI: 10.1088/0953-8984/14/35/301
- Erlebacher, J.; Aziz, M. J.; Chason, E.; Sinclair, M. B; Floro, J. A *Phys.Rev. Lett.*, **1999**, *82*, 2330.
- DOI: <u>10.1103/PhysRevLett.82.2330</u>
 Frost, F.; Schindler, A.; Bigl, F. *Phys. Rev. Lett.* **2000**, *85*, 4116.
 DOI: <u>10.1103/PhysRevLett.85.4116</u>
- Gago, R.; Vazquez, L.; Cuerno, R.; Varela, M.; Ballesteros, C.; Albella, J. M.; *Appl. Phys. Lett.* 2001, 78, 3316.
 DOI: 10.1063/1.1372358
- Ziberi, B.; Frost, F.; Höche, T.; Rauschenbach, B. Appl. Phys. Lett. 2005, 87, 033113.
- **DOI:** <u>10.1063/1.2000342</u> 6. Frost, F. ; Rauschenbach, B.; Ap
- Frost, F. ; Rauschenbach, B.; Appl. Phys. A: Mater. Sci. Process. 2003, 77, 1. DOI: 10.1007/s00339-002-2059-3
- Ziberi, B.; Frost, F.; Höche, Th.; Rauschenbach, B. *Phys. Rev. B* 2005, 72, 235310.
 DOI: 10.110.001
- **DOI:** <u>10.1103/PhysRevB.72.235310</u> 8. Facsko, S. ; Dekorsy, T. ; Koerdt, C.; Trappe, C. ; Kurz, H. ; Vogt, A. ; Hartangel H. L. Spinner **1000**, 285, 1551
- A.; Hartnagel, H. L. *Science* **1999**, *285*, 1551. **DOI:** <u>10.1126/science.285.5433.1551</u>
- Ziberi,B; Frost, F. ; Neumann. Tarah,M; Rauschenbach,B Appl. Phys. Lett. 2008, 92, 063102.
 DOI: 10.1063/1.2841641
- Macko, S.; Frost, F; Engler, M; Hirsch, D.; Höche, T.; Grenzer, J; Michely., T New J. Phys. 2011, 13 073017. DOI: 10.1088/1367-2630/13/7/073017
- Zhang, K; Rotter, F; Uhrmacher, M; Ronning, C; Krauser, J; Hofsäss, H New J. Phys. 2007, 9 29.
- **DOI:** <u>10.1088/1367-2630/9/2/029</u> 12. Hofsäss,H; Zhang,K *Appl. Phy. A* **2008**, *92* 517. **DOI:** <u>10.1007/s00339-008-4678-9</u>
- Yang, Z. Adv. Mat. Lett. 2011, 2, 195.
 DOI: 10.5185/amlett.2011.1212
 DOI: 10.1103/PhysRevB.80.155434
- Oates, T. W. H.; Keller, A.; Facsko, S; Muecklich, A. *Plasmonics* 2007, 2, 47. DOI:10.1063/1.2959080
- 15. Sarathlal, K.V; Kumar,D; Gupta,Ajay. Appl. Phys. Lett. 2011, 98, 123111.
 - DOI: <u>10.1063/1.3567731</u>
- Fassbender, J; Strache, T; Liedke, M O; Markó, D; Wintz, S; Lenz, K; Keller, A; Facsko, S; Mönch, I; McCord, J; *New J. Phys* 2009, *11*, 125002.
- DOI: <u>10.1088/1367-2630/11/12/125002</u>
 17. Sarathlal,K.V; Kumar, D.; Ganesan, V.; Gupta, Ajay. *Appl. Surf. Sci.* **2011**, *258*, 4116
- DOI: 10.1016/j.apsusc.2011.07.105
 18. Keller, A; Roßbach, S; Facsko, S; Moller, W. Nanotechnology 2008, 19, 135303.
 DOI: 10.1088/0957-4484/19/13/135303
- Alkemade, P.F.A; Jiang, Z.X; J. Vac. Sci. Technol. B 2001, 19(5), 1699.
- DOI: <u>10.1116/1.1389903</u>
 20. Bradley, R. M.; Harper, J. M. E. J. Vac. Sci. Technol.A **1988**, 6, 2390.
- DOI: <u>10.1116/1.575561</u>
 21. Koponen, I.; Hautala, M.; Sievanen, O.P. *Phys. Rev. Lett.* **1997**, 78, 2612.
- **DOI:** <u>10.1103/PhysRevLett.78.2612</u> 22. Cuerno, R.; Barabási, A.L. *Phys. Rev. Lett.* **1995**, 74, 4746.
- DOI:<u>10.1103/PhysRevLett.74.4746</u> 23. Sivashinsky, G. I.; Michelson, D. M. Prog. Theor. Phys. **1980**, 63,
- 2112. DOI: <u>10.1143/PTP.63.2112</u>
- Wittmaack, K. J. Vac. Sci. Technol.A 1990, 8, 2246.
 DOI: doi:10.1116/1.576744
- Vajo, J. J.; Doty R. E.; Cirlin, E.H. J. Vac. Sci. Technol. A 1996, 14, 2709.

DOI:10.1116/1.580192

- Rusponi, S.; Costantini, G.; Boragno, C.; Valbusa, U. Phys. Rev. Lett. 1998, 81, 2735.
 DOL 10.1102 (D. D. L. 10.1.2725)
- DOI: <u>10.1103/PhysRevLett.81.2735</u>
 27. Eklund, E. A.; Bruinsma, R. ; Rudnick, J.; Williams, R. S. *Phys. Rev. Lett.* **1991**, 67, 1759.
 DOI: 10.1102/DIF 75.
- DOI: <u>10.1103/PhysRevLett.67.1759</u>
 28. Yang, H.N.; Wang, G.C.; Lu, T.M. *Phys. Rev. B* **1994**, *50*, 7635.
 DOI: <u>10.1103/PhysRevB.50.7635</u>.
- 29. Chan, W.L; Pavenayotin, N; Chason, E; *Phy.Rev. B*, **2004**, *69*, 245413.
- DOI: 10.1103/PhysRevB.69.245413
 30. Rusponi, S.; Costantini, G.; Boragno, C.; Valbusa, U. *Phy. Rev. Lett.* 1998, *81*, 2735.
 DOI:10.1103/PhysRevLett.81.2735
- Costantini, G.; Rusponi, S.; Buatier de Mongeot, F.; Boragno, C.; Valbusa, U. J. Phys.: Condens. Matter 2001, 13, 5875. DOI: 10.1088/0953-8984/13/26/303
- Pelaz, L; Marqués, L.A; Barbolla, J. J. Appl. Phy 2004, 96, 5947.
 DOI: 10.1063/1.1808484
- 33. Gevers, P. M.; Gielis, J. J. H.; Beijerinck, H. C. W.; Van de Sanden, M. C. M; Kessels W. M. M. J. Vac. Sci. Technol. A 2010, 28(2), 293.
 DOI: 10.1116/1.3305812

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