Chapter 10

Summary

In this chapter, the studies presented in this thesis have been summarized.
In recent days, ion beam sputtering is established as a powerful technique to induce nanostructures on any kind of material surface i.e., metal, semiconductor or insulator. The reasons those makes this technique advantageous over other nanopatterning methods like lithographic techniques, MBE etc. are its fast processing time to induce large area surface nanostructuring by just simple exposure to ion beam. Apart from surface modification, IBS also affects the compositional, electrical, magnetic and mechanical properties of surface. In this thesis work, we have studied the surface modification of Si, Ge and GaAs by low-energy ion beam sputtering from topographical, compositional and microstructural aspects. The experimental results are discussed in context of different theoretical models of ion beam sputtering.

In case of low energy IBS study on Si, the role of ion incidence angle, ion sputtering time, ion energy, ion current density, substrate rotation and substrate temperature are discussed.

- Depending on ion incidence angle, Si surfaces show different topographies like ripples or irregular dots. For certain range of oblique ion incidences (55°-70°), wave-vectors of the ripple nanopatterns are aligned parallel to ion beam projection and after a certain critical angle >75°, the ripples are rotated orthogonally. In transition region 75°, the superposition of parallel and perpendicular mode patterns generates irregular mound structures. The theoretical models which are proposed to describe the ion beam nanopatterning are mostly based on linear BH theory where the curvature dependent ion erosion and surface smoothing mechanism are thought to play major role in creating surface instability during ion beam sputtering. While discussing the resultant topographies according to existing theories, we reached to an important conclusion that mass redistribution of surface adatoms plays dominant role in parallel mode ripple pattern formation rather than the curvature dependent ion erosion process.

- Next, we have studied the temporal evolution of parallel mode ripple patterns at 65° to get an insight into its dynamics during ion beam erosion process. At low sputtering time, ripple amplitude follows a power law behavior with the growth exponent $\beta = 0.27 \pm 0.02$. These initial ripple patterns are found to overlay by larger corrugation of another ripple like patterns at large lateral

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scales for higher fluences whose ripple wavevector is also aligned parallel to ion beam projection and ripple amplitude shows saturation. The dynamic scaling behavior at high fluence and short lateral scales indicates the presence of a KS like instability but at large lateral scales, exhibits different roughness exponents $\alpha_n = 0.63 \pm 0.05$ and $\alpha_p = 0.3 \pm 0.08$ in the direction normal and parallel to the ion beam projection respectively. The anisotropic scaling behavior does not agree with the prediction of KS equation, i.e., isotropic logarithmic scaling [103] or rotated ripple structures [221] which is expected for the present experimental conditions.

- We have also investigated the ripple morphological behavior as function of ion energy and ion current density individually i.e., when one is varied the other one is kept fixed. Experimental results show that the ripple wavelength and amplitude can easily and finely be tuned by those parameters. The quantitative estimation of ripple wavelength and its changes with ion energy and current density indicate the smoothing mechanism via ion induced viscous flow plays the major role for pattern formation on Si surface at room temperature.

- To study the role of substrate temperature on parallel mode ripple pattern on Si surface, the ion beam sputtering was performed at elevated temperatures from 20-450°C for low ($1 \times 10^{18}$ cm$^2$) to high fluence ($1 \times 10^{20}$ cm$^2$) regime. At low fluences, the ripple pattern is tend to diminish with increase of temperature. But at high fluence, the ripples show an orthogonal transition in its pattern orientation from the substrate temperature $\approx 350^\circ$ and its becomes prominent if the substrate temperature is increased more. We have also studied the temporal evolution of these rotated ripples at 450°C to investigate its dynamics. At initial sputtering time, the experimental results show almost smooth surface and some low amplitude grains. After a fluence of $2 \times 10^{19}$ cm$^2$, the perpendicular mode ripples are started to evolve. The discussion of temperature dependent experimental results in context of theoretical models of ion beam nanopatterning reveals that the ion induced viscous flow acts as a dominant smoothing mechanism during temperature induced pattern evolution process. By EDS analysis, it was confirmed that the temperature induced surface morphologies
are not induced by impurity or any kind of metal contaminations. On the other hand, cross-sectional TEM study shows that temperature leads the reduction in thickness of amorphous layer but enhances crystalline defects in near-surface region of ion sputtered surfaces.

- Next, we have studied morphological evolution as well as the dynamic scaling behavior of Si surface during grazing incidence (75°) Ar⁺ ion beam sputtering for various ion fluences at room temperature. Initially, the surface shows nanometer size dot-like structures which grow and evolve to cone/needle-like structures with increase of sputtering time, pointing towards the beam direction. The dynamic scaling behavior of fluence dependent surface patterns is discussed in terms of conventional and anomalous dynamic scaling theory. The scaling behavior shows a crossover at t = 4 min corresponding to the fluence 1.5 × 10¹⁸ cm⁻². Before fluence φₜ ≤ 1.5 × 10¹⁸ cm⁻², the estimated scaling component does not follow the scaling relation. This is identified as linear instability region where generally scaling exponents do not satisfy the dynamic scaling relation according to theories of ion beam nanopatterning. However, at higher fluences beyond φₜ, the system approaches to the asymptotic regime and the erosion dynamics follow super-rough scaling behavior with the set of exponents α = 1.6 ± 0.1, β = 0.99 ± 0.03, α_{local} = 1 and β_{local} = 0.63 ± 0.05. This super-rough scaling is associated with the instability due to non-local ion reflection effects that take place during the growth of conical patterns from the dot structures.

- Our next study is the investigation on the role of substrate rotation during low energy Ar⁺ ion beam sputtering on Si(100) surface at different oblique incidence angle. Experimental results show the formation of irregular isotropic nanodots at incidence angle of 65° as result of substrate rotation 5 rpm where periodic ripple patterns are generated without substrate rotation. Similarly, smooth surface and hexagonally ordered nanodots (isotropic structure) are respectively observed at 85° and 75° due to substrate rotation 5 rpm, where perpendicular mode ripples and irregular mound structures are formed in absence of substrate rotation. Then, we have examined the role of substrate rotation in the evolution of ordered nanodots from irregular mound structures at 75° angle of incidence.

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For 500 eV Ar$^+$ ion irradiation on Si surface, hexagonally ordered nanodots are produced just after 1 rpm. In context of dot’s amplitude, it shows a minimum at speed 2rpm. Surprisingly, after that it increases and saturates for higher rotation values. Cross-sectional TEM study of nanodots reveals that the core of the dots are highly crystalline and are not influenced by any metal impurities. The beam energy variation studies indicate the existence of a threshold energy above which the regular dot pattern appears. Finally, it is shown that the dot patterns obtained under rotated and off-normal ion beam sputtering can be described by the modified version of dKS equation in the limit of high rotation frequencies.

We have also studied the modification of Ge surface at elevated temperatures by hyperthermal (30 eV) ion irradiation. In general, the surface instability during ion beam sputtering is considered to arise due to interplay between curvature dependent ion erosion and smoothening via surface diffusion. For 30 eV ion bombardment on solid, as the impact energy is close to the lattice displacement energy, a very low sputtering yield and negligible ion erosion is expected. Here we show that the sputter erosion of target is actually not needed to induce surface instability for pattern formation. A dense array of faceted nanostructures evolves on the Ge(001) surfaces for normal incidence 30 eV ion bombardment at elevated temperatures. There is a narrow window of temperatures (225-350$^\circ$C), just above the crystal annealing temperature, in which the grown pattern resembles to the checkerboard pattern of alternating mounds and pits. The measured roughness exponent characterizing the interface morphology is found to be extremely sensitive to substrate temperature and follows the dynamics of diffusion-bias generated growth processes.

To extend the understanding of pattern formation mechanism for compound materials, we have performed the low energy ion beam sputtering on GaAs surface for different conditions. Ripple formation driven by Ehrlich-Schwoebel barrier is evidenced for normal incidence 30 eV Ar$^+$ bombardment on GaAs(001) surface at elevated target temperatures. The patterns follow the twofold symmetry of the bombarded crystal surface. The ridges of the ripples are found to align along the $\langle 1\bar{1}0 \rangle$ direction. The results are described by a non-linear continuum equation based on biased diffusion of
adspecies created by ion impact. On the other hand, highly ordered and defect-free nanoripples are formed on GaAs(001) surface at normal incidence when the sample temperature is kept above the recrystallization temperature and bombarded by 1 keV Ar\(^+\) ion beam. In this case, the ripple crests are also aligned along the \(\langle 1\bar{1}0\rangle\) direction. A coupled continuum equations, involving height and compositional variations and plugged with the diffusion bias, can reproduce remarkably well the experimental results.

From the above described summary of the systematic experimental studies, one can realize that ion beam sputtering can be considered as a potential and alternative approach for the production of large scale nanopatterning on single as well as on compound semiconductors. By tuning the sputtering condition in terms of different processing parameters like ion incidence angle, substrate temperature, substrate rotation etc., highly ordered ripples, dots or checkerboard patterns can be produced. The crucial thing is to identify the proper sputtering condition for that particular ion-target combination to obtain highly ordered patterns. One other important contribution by our work to the field of ion beam nanopatterning is the production of crystalline nanopatterns by ion beam sputtering. It is well-known that due to the ion beam sputtering, the upper surface of materials gets amorphized at room temperature which reduces the potential applications of nanopatterns. This had been a major issue with this technique for long time. This thesis work in a part of it presents pure and highly crystalline nanodots on Si surface at room temperature by 500 eV Ar\(^+\) ion sputtering at oblique incidence with simultaneous substrate rotation. Moreover, we have shown experimentally that the ion erosion is not necessary to generate patterns. This has been done by performing irradiation with 30 eV ion energies on Ge and GaAs surfaces at elevated temperatures. Near recrystallization temperature, both the surface shows highly ordered and crystalline nanopatterns \textit{i.e.}, checkerboard patterns for Ge and ripple patterns for GaAs surfaces.