Self-Organized Nanostructures on GaSb(001) Surfaces Due to Low-Energy Ion Irradiation

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Abstract:

Surface modification of GaSb(001) induced by low energy (2 to 4.5 keV) Ar ion beam irradiation has been investigated by means of atomic force microscopy (C-AFM). It has been found, that depending on the ion beam parameters, i.e. energy and fluence, narrow size distribution of nanostructure patterns have been developed on the irradiated surfaces. Particularly, at incidence angle of 80^o with respect to the surface normal, remarkably ordered ripple like structures are formed. The wave vector of the ripple pattern is oriented perpendicular to the direction of the ion beam projection on the irradiated surface. It has been also found, that the surface roughness, in terms of RMS, is initially a decreasing function of the ion fluence, but above medium fluence (of 1×10^{16} ions/cm^2) it increases with the irradiation time. The results are discussed in terms of ballistic processes of sputtering and existing theories describing the ion beam induced surface modification.

Keywords: surface patterning, ion irradiation, ripples, AIII-BV, atomic force microscopy.

Introduction :

Surface patterning on nanometer scale on crystalline and amorphous solids due to ion bombardment has become a topic of rapidly increasing interest [1]. In this regard, with the fast-growing interest in nanotechnology, an ion beam sputtering (IBS) is frequently used as an alternative process for the generation of various nanostructured surfaces or interfaces via self-organization processes. One reason is certainly the potential of such nanoscale patterns for various important applications in contemporary electronics, materials science, molecular biology and medicine. Regarding self-organization effects and spontaneous pattern formation during ion sputtering, the phenomenon of ripple formation on the irradiated surfaces was intensely studied for a last few decades[2]. The first surface structuring in form of periodic patterns was observed in the experiments by Navez *et al* in 1962 [3]. Since then, patterns have been found on a variety of materials irradiated with ion beams, such as metals, semiconductors and insulators [4–12]. The main mechanisms that determine the surface morphology of amorphous solids undergoing oblique ion bombardment is quite-well documented by Bradley and Harper (BH) approach [13].

According to BH theory, the initial modulation is given by the microscopic roughness of the surface and the stochastic nature of the sputtering process proposed by Sigmund [14]. The probability that an atom is removed from the irradiated surface is dependent on the deposited energy by all penetrating projectile ions. It implies that the trough regions are eroded faster than the crests which finally lead to the destabilization of the surface topography and increase of the surface roughness. This instability process competes with a smoothing mechanism, e.g. surface diffusion or viscous flow, both being of thermal [15] or ion bombardment origin [16]. The interplay between these two processes, the ion-induced surface instability and surface relaxation processes determine a characteristic wavelength of the created surface pattern [13]. However, BH approach was not sufficient to explain many of experimental observations, thus several modifications of the BH model have been proposed to explain, for example, the pattern stabilization for long sputtering times [17], nanodots formation for the isotropic case [18] and, more recently, the observation of ordering and coarsening [19,20].

A variety of nanometer-scale topographic features in form of dots, ripples or pillars is observed on the ion irradiated compound III-V semiconductor surfaces. For this class of multi-component targets, like for example GaSb, the ion bombardment, apart from morphological evolution, can also induce changes in the elemental composition of the irradiated surfaces. Facsko *et al*. investigated low-energy, normal incidence Ar^+ sputtering of GaSb (100) surfaces and observed that, as the erosion proceeds, self-organized nano-scale dots develop on the surface [21]. Y. Homma observed filament-like microtextures on GaSb implanted with Cs^+ ions at low energy [22] and Nitta and Taniwaki observed the formation of a cellular structure in the irradiated GaSb and InSb crystals, due to a diffusion of the point defects induced by ion implantation [23, 24]. A. Lugstein *et al.* observed a sponge-like structure built up of fibers with encapsulated GaSb nanocrystallites on the GaSb (100) surface after exposure to 50 keV focused Ga^+ ion beam. They explained the fibers formation based on the idea of a catalytic vapour liquid solid (VLS) mechanism [25]. According to VLS approach, a metal droplet catalyzes the decomposition of a precursor gas, which plays a role of a nanowire material reservoir by eutectic liquid formation, and finally leads to a precipitation of the nanowire due to a super saturation. More recently, S. Le Roy *et al.* [26] have observed a pillar growth on GaSb during the very low energy (500 eV) ion beam irradiation. They ascribed the growth of the pillars to gallium segregation on the irradiation surface.

In the present work, the nanostructures formation on GaSb (001) surface have been studied as a function of various beam parameters i.e. energy and fluence of the Ar^+ ion beams at room temperature.. Depending on the sputtering parameters different kind of nanostructures like dots, big droplets and ripples have been observed by using atomic force microscopy (AFM) in contact mode in air.

Experimental:

Epi-ready, polished GaSb(001) wafers, purchased from Kelpin Crystals (Neuhausen, Germany) were used in the experiments as a substrate. All samples, with sizes of 5x5 mm^2, were tightly mounted on a molybdenum stage plate (fixed on a copper holder) with the tungsten clamp. The *ex situ* pre-cleaning has been done with propane and blowing by pure nitrogen gas to remove the residual contaminant occurring due to the cutting of the wafers. The *in situ* sample cleaning has been done in the preparation chamber following a standard procedure of annealing to 450 K for 2 h. The sample temperature was measured with Chromel-Alumel thermocouple fixed on the sample holder.

The surface nanostructuring has been carried out with the broad, defocused Ar^+ beam using the plasma ion source. The ion beam energy E_{ion} in the range from 2 keV to 4.5 keV was used. The average ion current density, depending on the projectile energy, was about 60-100 $\mu A/cm^2$. The estimated projectile fluence was varied between $1x10^{15}$ to $2x10^{18}$ ions/cm^2. Followed the nanostructuring process, the samples were taken *ex situ* for AFM measurements. The resulting surface topography has been analyzed by the atomic force microscopy in contact mode (c-AFM) with the CP Park Scientific Instruments microscope using silicon cantilevers with a nominal tip radius of 20 nm. The average ripple wavelength was measured directly from AFM topography images, as well as from the corresponding 2D height–height autocorrelation functions. For better statistics, the AFM measurements have been done at several different areas of the irradiated sample surface.

Result and Discussion:

AFM topographic image of a pristine GaSb (001) surface is presented in Fig. 1. The surface is flat, with a RMS roughness of about 0.6 nm and some randomly spread pits of depth in the range of about 1nm are seen.

The ion energy dependent ripple formation on GaSb (001) surface at 80^o off-normal incidence angle and the fluence $4x10^{17}$ ions/cm^2 is shown in Figure 2. The ripples are covered with dots of different sizes. The periodicity (wavelength) of the ripples and the dot size depend on incidence ion energy as is shown in Fig.2. The ripples are oriented along the ion beam projection on the surface.

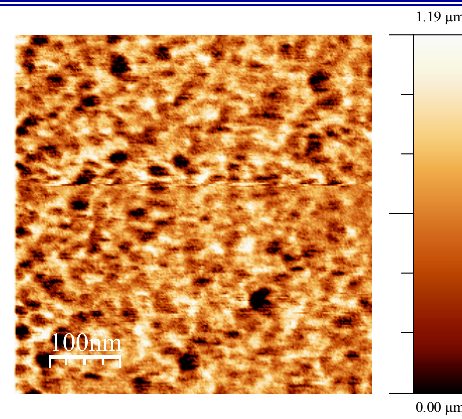

Fig.1. AFM image of unbombarded GaSb(001).

From Fig.3. the fluctuation of the ripple wavelengths around 175 nm has been observed and in general, the RMS roughness decrease with ion energy. It attributes to the fact that with the increasing energy of projectile, the smoothening processes take over the roughening (erosion) simply due to the deeper energy deposition in the sample.

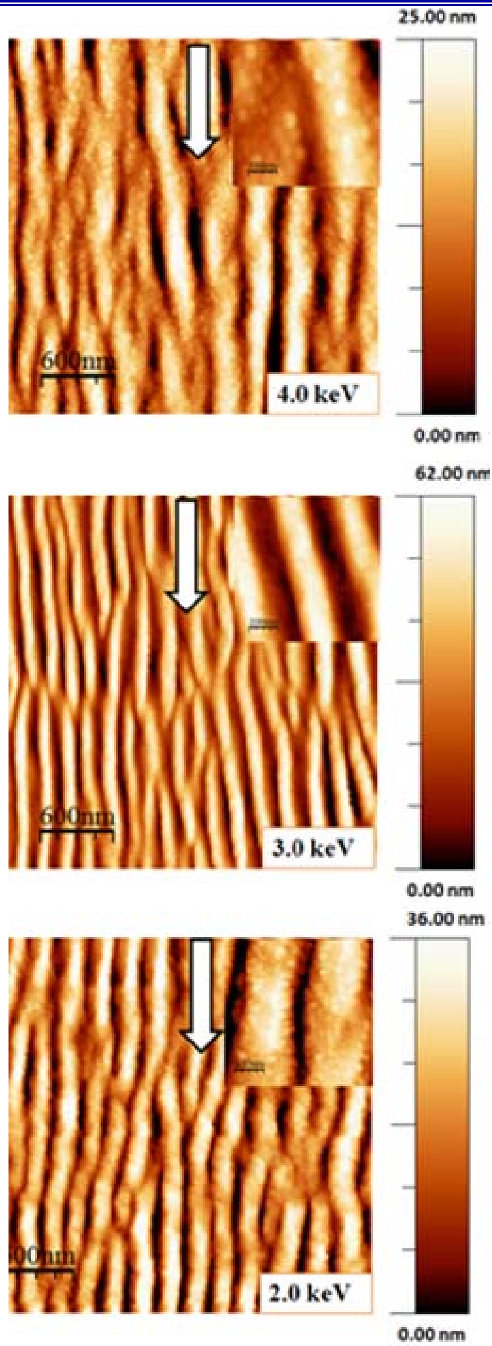

Fig. 2. AFM topography images (3x3 μm^2) of GaSb(001) surfaces irradiated with Ar ion beam at the energy of (a) 2.0 keV, (b) 3.0 keV and (c) 4.0 keV. The modification was performed at room temperature with the incident angle of 80^o off-normal and ion fluence of 4x10^{17}ion/cm^2. In the insets high resolution images are shown.

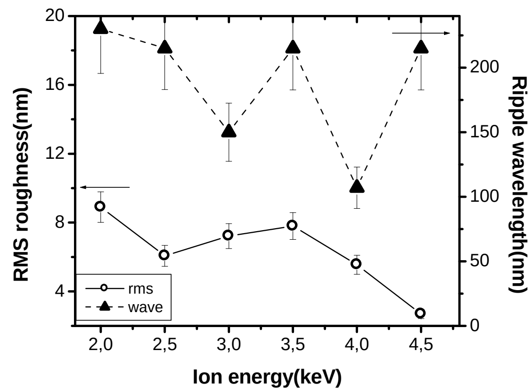

Fig.3. Ripple wavelength and RMS roughness of the GaSb(001) surface as a function of the Ar ion beam energy.

In Fig. 4. AFM topography images of GaSb(001) bombarded by Ar ions at angle of 80^o and energy 2 keV as a function of the ion fluence is shown. For the low ion fluence (1.7 x10^{16} ions/cm^2, Fig. 4a) the separated and elongated dot-like features on the irradiated surface are seen. They are arranged along the ion beam direction and serve as a nucleus of the ripples which develop for higher fluence (Fig. 4b). Highly ordered ripples are formed at still higher ion fluence (3.5x10^{17} ions/cm^2, Fig. 4c). These ripples are covered with dot-like structures of different height, from 4 to 10 nm, and width, from 10 to 35 nm. The arrows indicate the ion beam direction on the surfaces and the images at the center are of 600 x 600 nm^2 size.

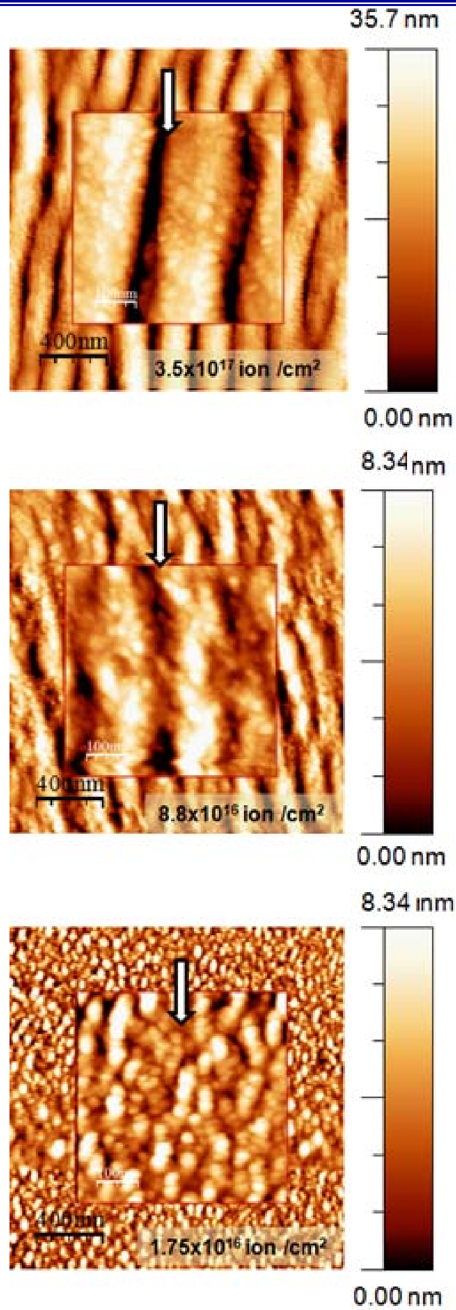

Fig. 4. AFM topography images (2x2 μm^2) of GaSb(001) surfaces irradiated with Ar ion beam at different fluences of (a) 1.7 x 10^{16} ions/cm^2, (b) 8.8 x 10^{16} ions/cm^2 and (c) 3.5 x 10^{17} ions/cm^2. The modification was performed at room temperature with the incident angle of 80^o off-normal, ion beam energy of 2 keV. The arrows indicate the ion beam direction on the surfaces and the images at the center are of 600 x 600 nm^2 size.

In Fig. 5, the measured RMS as a function of the Ar ion beam fluence (ϕ), at the incidence angle of 80^o and the ion energy of 2.0 keV, is shown. In the range of the ion fluences studied, the RMS roughness follows the power scaling relation $RMS \sim \phi^{\beta}$ with the growth exponent $\beta = 0.82$.

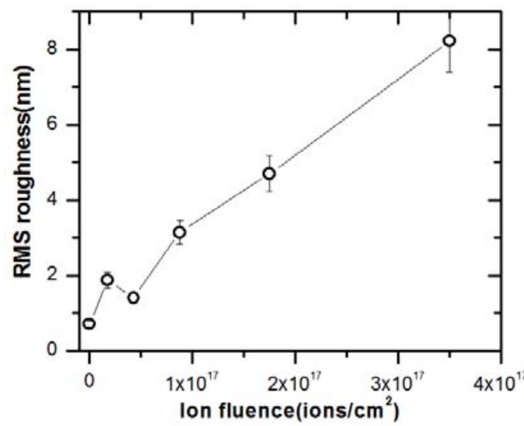

Fig.5. Variation of the RMS roughness and ripple wavelength with ion fluence (001). Bombarded by Ar ion at 80^o off-normal, ion energy 2keV.

According to the BH model, the amplitude (and correspondingly the RMS roughness) of the periodic structures due to ion bombardment should depend exponentially on the irradiation time, a quantity equivalent to the ion fluence. However, the observed scaling law $RMS \sim \phi^{\beta}$ disagree with the predictions of the BH model. As demonstrated theoretically, a power law scaling $RMS \sim \phi^{\beta}$ is a consequence of various nonlinear effects occurring on the irradiated surfaces [9]. In the present case, it is plausible to assign the observed scaling law behaviour of modified GaSb(001) surface with the diffuse-interface model for composition-induced nanopatterning during ion bombardment proposed by S. Le Loy *et al.* [30]. According to this model a simultaneous segregation of the sample components and shielding mechanism is based on the surface enrichment in gallium due to the big difference in sputtering yield of Ga and Sb components. Above a critical concentration, Ga segregates to form a cap which acts as a sputtering shield. Since the difference in sputtering yield as well as the tendency for segregation is properties of the bulk material, the Ga-rich shield is resupplied during the ion sputtering. The change in surface composition develops faster than the BH instability, with a smaller characteristic wavelength, and leads to the formation of Ga-rich "droplets" as well as unstable surface. With the increasing ion fluence the surface pattern develops by taking into account the difference in sputtering yield between Ga and GaSb. The excess Ga, continuously replenished by bulk GaSb, enter the droplets which could elongate due to attachment of droplets to each other. With the increasing fluence the difference of sputtering yield between Ga and GaSb causes the surface roughening and may be the origin of the ripple development at grazing incident angle.

Conclusion:

In this paper, the role of the ion energy and ion fluence on the formation of self-organized nanostructures on GaSb(001) surfaces during Ar ion bombardment at room temperature have been summarized. Depending on the experimental conditions variety of nanostructures are observed. At low fluence, dots and big droplets are developed on the irradiated surface. Furthermore, the ripples develop with wave vectors normal to the incident ion beam on the irradiated surface.

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