Nuclear Instruments and Methods in Physics Research B 267 (2009) 2752–2756

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

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

Temperature-dependent surface modification of InSb(0 0 1) crystal by low-energy ion bombardment

S.R. Saeed [1], O.P. Sinha [2], F. Krok, T. Zembok, R. Pedrys *, M. Szymonski

Marian Smoluchowski Institute of Physics, Jagiellonian University, Reymonta 4, 30-059 Krakow, Poland

ARTICLE INFO

Article history:
Available online 27 May 2009

PACS:
79.20.Rf
34.35.+a
82.80.Rt

Keywords:
Nano-patterning
Ripples
Surface modification
Ion sputtering
Ion-induced diffusion
Time-of-flight-SIMS
InSb

ABSTRACT

Structural and compositional modification of InSb(0 0 1) single crystal surfaces induced by oblique incidence 2–5 keV Ar and Xe ion irradiation have been investigated by means of scanning tunneling and atomic force microscopies, and time-of-flight mass spectroscopy of secondary ion emission. In general, ion-induced patterns (networks of nanowires, or ripples) are angle of incidence- and fluence-dependent. Temperature dependences (from 300 to 600 K) of the RMS roughness and of the ripple wavelength have been determined for the samples bombarded with various fluences. Secondary ion emission from an InSb(0 0 1) surface exposed to 4.5 keV Ar^+ ions has been investigated with a linear TOF spectrometer working in a static mode. Mass spectra of the sputtered In^+, Sb^+ and In_2^+ secondary ions have been measured both for the non-bombarded (0 0 1) surface and for the surface previously exposed to a fluence of 10^{16} ions/cm^2. In^+ and In_2^+ intensities for the irradiated sample are much higher in comparison to the non-bombarded one, whereas Sb^+ ions show a reversed tendency. This behavior suggests a significant In-enrichment at the InSb(0 0 1) surface caused by the ion bombardment.

1. Introduction

Surface patterning at nanometer length scale has received considerable attention due to its potential applications in various fields of research and technology [1]. Ion beam techniques are of particular interest due to their possibilities for the creation of large area patterns, and versatility in adjusting the pattern characteristics by properly tuning the beam parameters. Some of such appealing morphological developments are known as nanodot and nanoripple formation on various irradiated surfaces [2–19].

Recently, ion-induced nanostructuring of III–V semiconductor surfaces has been investigated extensively, since it shows different surface morphologies depending on ion energy, flux, fluence, and ion species. For example, normal incidence Ar^+ irradiation of GaSb surfaces results in the formation of crystalline dots, 35 nm in diameter, arranged in a regular hexagonal lattice, whereas under off-normal bombardment with 420 eV ions, coherent ripples are formed with a period of 35 nm [20]. More recently Frost et al. [21] reported that a similar array of close-packed mounds could be obtained by oblique incidence Ar^+ ion sputtering of an InP surface with a simultaneous rotation of the sample. The characteristic pattern wavelength increased with the sputtering time following a simple scaling law: $\lambda \sim t^\gamma$ with $\gamma \cong 0.26$. This last work has been extended for other compound semiconductors, such as GaSb, InSb and InAs [12] with essentially the same observations. Evolution and coarsening behavior of self-assembled nanodots fabricated on an InP surface by 3 keV Ar ion sputtering have also been studied by Parmanik et al. [22] in a slightly off-normal geometry but in the absence of rotation. For small sputtering durations, the dots coarsen and agglomerate, up to a critical time, beyond which an inverse coarsening, fragmentation of dots and a smoothed surface are observed.

In most of the papers cited above, surface morphology was examined with an ambient scanning force microscopy (AFM) performed "post mortem", i.e. exposing the sample in air after ion sputtering. Usually, the surface composition during the ion bombardment was not monitored, assuming that the surface composition is stable at dynamic equilibrium conditions. Ion sputtering of compound semiconductors, however, often leads to surface composition changes due to preferential sputtering of one component (usually BV) over the other [15]. Therefore, the knowledge of irradiated surface composition and its potential effect on surface morphology is of the utmost importance.

In the present paper, we report on surface morphology modifications of an InSb(0 0 1) crystal induced by low-energy

* Corresponding author.
E-mail address: ufpedrys@cyf-kr.edu.pl (R. Pedrys).
[1] On leave from Department of Physics, Sulaimani University, Sulaimani, Kurdistan-Region, Iraq.
[2] Visiting Scientist from the Amity Institute of Nanotechnology, Amity University, Uttar Pradesh, Noida 201303, India.

0168-583X/$ - see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.nimb.2009.05.023

Ar^+ bombardment at various beam parameters and temperatures of the sample. The morphology measurements have been performed either under UHV conditions with STM/AFM system interconnected to the irradiation chamber, or in air with a contact mode AFM. Surface compositional information has been obtained with a linear ToF-SIMS spectrometer working in a static mode.

2. Experimental details

The surface morphology modification experiments are performed in an ultrahigh vacuum (UHV) system which consists of three chambers (sample preparation, surface characterisation and SPM imaging) with the base pressure in the range of 10^{-10} mbar. The InSb(0 0 1) epi-ready wafers, from Kelpin Crystals (Neuhausen, Germany), with sizes of 5×5 mm^2 are clamped to a molybdenum plate with the help of tungsten wires. The molybdenum plate is supported on the copper holder which can be heated by a resistor heater fixed inside the copper holder. The fresh sample is preannealed at 695 K for several hours. In order to get well-defined starting conditions, the InSb sample is further sputter-cleaned at 695 K by Ar ion bombardment for 1 h (using a 0.7 keV energy ion beam, current about 1.5 μA, rastered over 2 cm^2, incidence angle of ±60^o off-normal) for several cycles with subsequent annealing at the same temperature for 5 h. This procedure provides a clean and well-ordered surface showing a c(8×2) LEED pattern. For surface modification of such prepared samples we use ion bombardment with a focused Ar^+ beam (the diameter of the beam spot below 1 mm) scanned over the entire sample area at temperatures ranging from room temperature (RT) to 600 K. The temperature is measured with a chromel–alumel thermocouple fixed on the copper holder. The other experimental parameters could be varied too, i.e. the angle of incidence from 0° to 75° off-normal, the ion energy from 2 to 4.5 keV, and the ion fluence up to 4×10^{18} ions/cm^2. The average ion flux is kept between 16 and 18 μA/cm^2. The imaging of surface topographies is performed by atomic force microscopy in contact mode (C-AFM) under ambient conditions using a CP Park Scientific Instruments AFM microscope. Silicon nitride cantilevers with a tip of nominal radius in the range of 10 nm are used. The average wavelengths of the surface nanostructures are determined directly from AFM topography profiles as well as from 2D self-correlation functions of AFM images. For better statistics, the AFM measurements are taken at different places on the irradiated sample surface. For comparison, some images of irradiated surfaces are taken in UHV, i.e. the samples are directly transferred into the UHV STM/AFM chamber without exposing them to air. In this last case scanning probe microscopy is performed at room temperature with a Park Scientific Instruments VP2 AFM/STM. Tungsten tips (electrochemically etched) are used for the STM measurements.

Secondary ion mass spectrometry (SIMS) analysis of InSb(0 0 1) epi-ready wafers is performed with another apparatus which consists of a collision chamber equipped with ion and electron guns, QMS spectrometer, ToF-SIMS system and a UHV mechanical sample transfer mechanism by which investigated samples are introduced into the collision chamber. In the chamber, the wafer is placed on a moveable holder, kept at room temperature. A 4.5 keV Ar^+ ion beam is focused onto an InSb sample under an angle of incidence of 45^o off the surface normal. The ion current is measured by turning a Faraday cup into the beam, in front of the surface. The base pressure in the target chamber is about 5×10^{-10} mbar, slightly increasing during the ion gun operation. Secondary positive ions ejected from the target are accelerated and mass analyzed by a linear ToF-SIMS analyzer. To ensure a static regime of secondary ion emission, 0.5–1.5 nA ion beam is chopped at a 4 kHz frequency with a pulse width below 50 ns. All spectral measurements are completed before a dose of about 10^{10} ions/cm^2 is reached.

3. Results

AFM images of InSb(0 0 1) surfaces irradiated with an Ar^+ beam at different temperatures are shown in Fig. 1. At room temperature (Fig. 1(a), image top left), the morphology of the irradiated surface consists of a set of straight, separated and elongated nanowire-like structures with a length up to a few micrometers. The wires are parallel to the projection of the ion beam on the irradiated surface and they are non-uniform in size along their length. Typically, they are extending from the dot-like "head" having an average diameter between 20 and 30 nm. With increasing temperature, considerable changes in surface morphology are seen (Fig. 1(b)–(d)). In the range from 400 to 500 K a ripple structure is clearly seen with a well-defined wavelength and small variations in height (in the range of a few nanometers). For the highest temperatures, above 570 K, a smooth and wavy surface morphology is observed (Fig. 1(d), image bottom right).

The dependences of the ripple wavelength on the target temperature taken for Ar and Xe ion bombardment are shown in Fig. 2. First, the ripple wavelengths seems to be independent of the ion species. Second, two different regions of the temperature dependence could be distinguished. These regions could reflect a different balance of various erosion and smoothing mechanisms taking place at low and high irradiation temperatures of the sample.

RMS surface roughness obtained for Ar and Xe ion irradiation versus the target temperatures is shown in Fig. 3. The RMS surface roughness for well-developed nanowires at RT is about 12 nm and at first it decreases with the increasing temperature and then stabilizes at the value of about 1 nm for temperatures above ≅375 K. It is seen that the target temperature at which surface roughness is stabilizing for each of the projectiles differs by about 50 K, corresponding to the change in surface morphology from nanowires to ripple-like structures.

ToF-SIMS signal intensities obtained for a non-bombarded InSb wafer and the one irradiated with 10^{16} ions/cm^2 are shown in Fig. 4.

4. Discussion

The standard theoretical approach, describing formation of the periodic pattern on the irradiated surfaces, refers to the linear theory of sputtering [23], and it is known as Bradley and Harper (BH) theory [24]. According to BH theory, the ripple formation during ion bombardment of amorphous solids is caused by the interplay between two counteracting effects, surface roughening and surface smoothing processes taking place on the irradiated samples. The time evolution of the roughening process is governed by a surface curvature-dependent ion sputtering, whereas the smoothing process originates from thermally activated diffusion [24], or ion-activated processes of material transport on the irradiated surfaces [25]. BH theory has been tested over a broad range of ion bombardment parameters such as the ion mass, the ion initial energy and the irradiation fluence. In several cases the model successfully predicts the ripple wavelength and orientation. On the other hand, BH theory is unable to explain a number of experimental features, such as saturation of the ripple amplitude [7,26], the appearance of rotated ripples [27–29] and kinetic roughening [30,31]. An extension of BH theory [32–35] has been made by including a noise term which accounts for the fluctuations caused by a stochastic nature of the sputtering process, and addition of a linear term which accounts for a tilt dependence of the sputtering yield, hence preventing the ripples from growing indefinitely. The nonlinear term leads to saturation of the surface roughness starting at an irradiation time called a "crossover time" [35]. Up to the crossover time, the BH linear term is sufficient to describe the surface topography. For longer irradiations, the nonlinear terms are required to