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Depth resolution enhancement by combined DSIMS and TOF-LEIS profiling

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ABSTRACT

A combination of dynamic secondary ion mass spectroscopy (DSIMS) and time-of-flight low-energy ion scattering (TOF-LEIS) has been applied to acquire a composition depth profile of MoSi multilayers. During the sequential Ar^+ sputtering secondary ions were monitored while in-between the sputtering cycles the TOF-LEIS spectra of scattered He neutrals were acquired. All the measured TOF-LEIS spectra versus sputtering time were displayed in one bitmap from which the depth profiles for different scattering depths were derived and analyzed. Analyzing the TOF-LEIS spectra of He particles scattered from the areas below the layer altered by ion-beam mixing led to an improvement of the depth resolution. In this way the resolution limits due to mixing phenomena can be overcome. Finally, the direct comparison of the DSIMS and TOF-LEIS depth profiles was carried out.

1. Introduction

Dynamic secondary ion mass spectroscopy (DSIMS) is an ion-beam sputtering technique frequently used for the depth profiling of elemental composition of thin films. It is generally accepted that DSIMS depth resolution is limited mainly by atom mixing. In order to reduce the thickness of an atom mixing region (an altered layer), the energy of primary ions has to be reduced to hundreds of eV. Even for these energies the thickness of the altered layer exceeds one monolayer. In addition to a pure ballistic mixing, there are another physical and chemical processes affecting depth resolution, i.e. diffusion, segregation, formation of new compounds, implantation, and mass back flow to the surface. These processes take place mainly inside the altered layer. Nevertheless, minimizing the thickness of the altered layer by lowering the ion beam energy is nowadays a major approach to meet the sub-nanometer depth resolution. This extremely difficult task can be only achieved using primary energies as low as 100 eV [1]. However, a monolayer depth resolution has not been achieved by sputtering techniques yet. The main advantage of SIMS is its high sensitivity in comparison to other available techniques. On the other hand, the intensity of detected secondary ions is strongly influenced by an ionization process taking place during sputtering, which makes a quantitative analysis difficult or even impossible. That is why the SIMS generally belongs to semi-quantitative methods.

Quantitative depth profiling is more straightforward when ion scattering is employed instead of sputtering (Rutherford backscattering – RBS, medium- and low energy ion scattering – MEIS, LEIS). All these scattering techniques are generally less sensitive than DSIMS. Depth resolution typically decreases with increasing energies of projectiles, but even for RBS where high energetic particles are used, a monolayer depth resolution can be achieved at the near surface region using ultra-high-depth resolution facilities [2,3]. In the case of time-of-flight LEIS (TOF-LEIS) even a sub-monolayer depth resolution at a surface may be achieved [4]. Moreover, a TOF-LEIS instrument can be regarded as low cost in comparison with MEIS and RBS. As there is an increasing demand to analyze deeper regions without losing the monolayer depth resolution, this cannot be satisfied by a standalone ion scattering technique since the depth resolution deteriorates at deeper regions due to energy loss straggling. This occurs during the penetration of particles into the matter and is caused by statistical fluctuations of the energy transfer in the collision processes. Fluctuations in the transfer of impacting energy to electrons and to nuclei are major contributors to the straggling. Broadening due to a finite detector solid angle, finite beam spot size, etc. can significantly affect depth resolution [5–7].

A feasible way to overcome the increasing energy straggling with depth is the removal of a surface layer by sputtering. In this way the region of interest gets closer to the surface and scattered ions undergo fewer collisions. The depth resolution in layers far from the surface in high resolution RBS may be improved by employing grazing incidence angle sputtering [8]. A depth resolution of 1.5 nm at deeper regions was also achieved by high resolution RBS, once the surface region was sequentially removed

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by 1 keV Xe^+ ions [9]. Besides the MEIS technique mentioned above, ESA-LEIS experiments (LEIS using an electrostatic analyzer) have been also used in combination with sputter depth profiling [10]. This technique was also used for a study of ion beam induced mixing during a sputter depth profiling of tantalum and lead marker layers in silicon [11]. Since ESA-LEIS detects only ions, its sampling depth is controlled by a particular neutralization process occurring during the ion scattering. In most cases it is limited to a few upper atomic layers of a surface [12]. The re-ionization probability of scattered particles depends on the atomic species presented at the sample surface and is especially large for oxygen. In case of presence of oxygen at the top most layers the detected scattered ions also come from subsurface layers as well and can be used for a nondestructive depth profiling. Diffusion and interaction studies with a depth resolution 0.2–0.1 nm and a maximum sampling depth of 5–10 nm were shown in [13]. In case of SIMS more than 90% of the secondary ions are coming from the outermost monolayer. That is why the information depth in ESA-LEIS depth profiles (no oxygen on the surface) is similar to SIMS. The sampling depth in the case of TOF-LEIS, where neutrals are detected as well, is given by the stopping power and energy of impinging ions. These neutrals are not affected by ionization effects depending on an elemental composition of the surface. For energies in the keV range the sampling depth is in the order of nanometers [14]. Hence, TOF-LEIS is suitable to measure the composition not only within the altered layer where the compositional information is degraded by ion induced mixing, but also beyond this layer. In this way the measurements of the composition by TOF-LEIS can provide better depth resolution than those obtained by DSIMS.

When sputtering is employed in combination with TOF-LEIS the secondary ions emerging during this process can bring additional information and enhance the sensitivity of the analysis. Thus the combination of DSIMS and TOF-LEIS becomes mutually beneficial and can overcome the above mentioned mixing and straggling limitations. In this paper we demonstrate an advantage of the simultaneous application of DSIMS and TOF-LEIS in analysis of ultra-thin MoSi layered structures being at physical limits of the SIMS method itself.

2. Experimental

A 3×(Mo/Si)/Mo ultra-thin multilayer structure (further called the MoSi sample) was deposited on a Si (1 1 1) substrate covered with a native oxide by rf magnetron sputtering at the Institute of Scientific Instruments, Czech Academy of Sciences in Brno. This technology has been developed for the commercial custom-made fabrication of X-ray interference mirrors. All measured samples were cut out of a 4 in. Si wafer on which the deposited multilayer was deposited. Before deposition, no treatment of the wafer was carried out and a planetary manipulator has been used in order to maximize the multilayer homogeneity. The base pressure in a deposition chamber equipped by a turbo molecular pump was 1×10^{-4} Pa. Deposition was carried out at the pressure 0.1 Pa in an argon atmosphere at an rf power of 100 W at room temperature. The purity of Si and Mo targets was 99.9995% and 99.9%, respectively. The samples were analyzed by SIMS, TOF-LEIS and High Resolution Transmission Electron Microscopy (HRTEM). The last technique was used for the calibration of depth profiles. The images were acquired at electron beam energy of 200 keV using the JEOL 2010 microscope providing a point-to-point resolution of 0.194 nm. The HRTEM cross-sectional specimen was prepared by mechanical polishing followed by ion-beam milling.

Since the standalone TOF-LEIS spectrometer used in our experiments has been described elsewhere [15] only a brief summary including additional changes providing sequential LEIS and DSIMS measurements is given here. The experiments were performed in an ultra high vacuum chamber with a base pressure of 8×10^{-8} Pa. An electron-impact ion-beam source (Omicron ISE 100) was used to produce He^+ ions in an energy range of 0.5–5 keV. The beam spot diameter at the sample surface was not larger than 1 mm. Ion pulses were generated by chopping the beam over a small aperture (1 mm in diameter). The maximum time resolution of the spectrometer was estimated to be 50 ns, but during the TOF-LEIS depth profiling the resolution was degraded to 150 ns to provide a sufficient intensity of the detected signal. After scattering from the sample surface (incident angle 18° with respect to the sample surface, scattering angle 152°) the particles were time separated in a 1105 mm long drift tube and finally hit a detector.

For the SIMS measurements an additional ISE 100 ion-beam source and a QMS 421 quadrupole mass analyzer (Balzers-Pfeiffer) were used. The ISE 100 was operated at low energies. The total pressure in the chamber was kept at 9×10^{-6} Pa. Sputtering under different impact angles (only Fig. 3) was provided by Ar^+ primary ions at energy of 500 eV. The current at the sample was approximately 0.2 μA. The spot size was ~0.3 mm and the area of the milled crater was ~15 mm^2. Computer controlled scanning of the beam was used to compensate for alterations in beam positioning caused by oblique incident angles. Electronic gating was also controlled by a computer. A plan view of the ion sources and analyzers is shown in Fig. 1. Sequential sputtering was done using Ar ions with an impact angle of 45° (with respect to the surface normal) at an energy of 800 eV and sample current 0.3 μA.

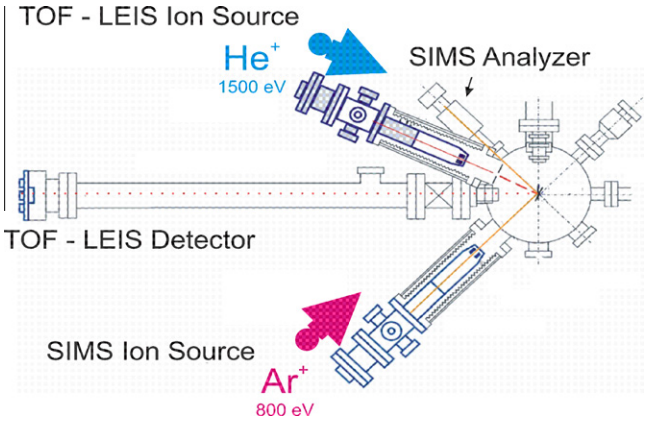

Fig. 1. Experimental setup for the combined DSIMS and TOF-LEIS techniques. The angle between the SIMS ion source and the mass analyzer – 90°, the angle between the longitudinal axis of the TOF-LEIS ion-beam source and the TOF-LEIS detector – 152° (scattering angle). The angle between the longitudinal axis of the TOF-LEIS ion-beam source and the horizontal plane of the experimental setup – 24°.

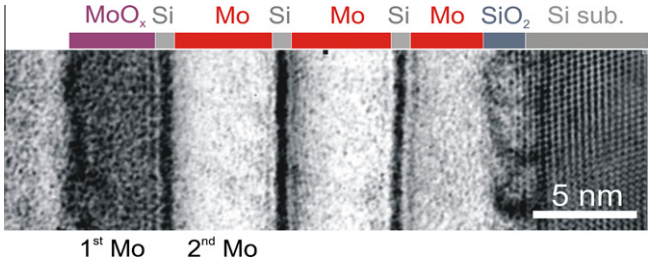

Fig. 2. Negative HRTEM image of 3×(Mo/Si)/Mo on a Si substrate.