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On the use of H$^+$ and Ar$^+$ ions for high spatial resolution depth profiling

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We report on the destructive sputtering of Ni–C multilayer coatings which have a periodicity of 2.7 nm, using H$^+$ as well as Ar$^+$ ions of 750 eV energy. The influence of ion bombardment on interface roughness, interface mixing and density has been investigated by monitoring this process in situ *using a soft X-ray reflection system. Ion bombardment has been demonstrated to decrease the roughness of a sputtered surface while a considerable intermixing of the interface below the sputtered layer occurs. Changes in the film density have not been observed.*

Introduction

Destructive sputtering with ions having an energy up to 2 keV in combination with a surface sensitive technique is widely used to obtain depth information on the chemical composition of thin films. Using this technique for the investigation of multilayer systems, the increasing microscopic roughness of the sputtered surface is reported to be a limiting factor for the depth resolution[1–3].

Ion impact is also used to modify thin films, as applied in Ion Beam Assisted Deposition (IBAD). Interesting effects like densification of the layers and improvement of interface roughness are expected[4].

Destructive sputtering of Ni–C multilayer coatings using Ar$^+$ and H$^+$ ions has been performed in order to investigate ion milling as a technique for depth profiling and ion beam modification of the coatings.

The soft X-ray reflection technique provided us with a tool to monitor both layer growth and sputter rate and appeared to be powerful in observing changes in the material density and interface roughness[5]. The computer program TRIM86 was used to simulate the sputter process[6].

Experimental

Deposition and sputtering have been performed in an uhv system with a base pressure of 10^{-8} Pa. Layers of carbon and nickel were deposited using two e-beam evaporators. Changes in layer thickness during growth and ion milling have been monitored by the soft X-ray reflection system ($\lambda = 3.16$ nm, $\theta = 35°$). A quartz crystal monitor could be used during deposition to compare layer thickness with the interference maxima in the reflected soft X-ray signal. The reflectivity as a function of film thickness was measured and fitted to a computational scheme, which solves the Fresnel equations and uses the Debye–Waller factor to describe interface roughnesses. The complex indices of refraction were calculated from the density and the atomic scattering factors f_1 and f_2[7]. A Kaufman source is used to produce H$^+$ or Ar$^+$ ions of 750 eV at an angle of incidence of 45°. The source was differently pumped, in order to keep the working pressure below 10^{-5} Pa.

Since a low surface roughness is needed to obtain a maximum X-ray reflectivity Si(111) crystals are used as substrates. The substrates were kept at room temperature.

Results and discussion

In Figure 1 the X-ray reflectivity during growth (solid line) and sputter process (dotted line) of a thick Ni layer on top of a Si(111) substrate is shown. The maximum thickness was 12.5 nm. The distance between the interference maxima (2.7 nm) extracted from the Bragg relation, agreed with the quartz crystal within its accuracy (3%) assuming bulk density for Ni. Simulation of the reflectivity curve during growth resulted in an increase of the surface roughness. Assuming an initial roughness of the Si surface of 0.4 nm, a final roughness of 0.8 nm was obtained. Sputtering has been performed using Ar$^+$ ions of 750 eV energy. From the sputtering time between two reflection maxima (2.7 nm) and the ion current (1.5×10^{-5} A cm^{-2}) a sputter yield of ≈ 1.5 Ni atoms ion^{-1} has been obtained. This is in good agreement with experiments from Fert *et al*[8]. The increasing amplitude of the oscillations and medium height of the reflected X-ray signal indicate either a sputter enhanced smoothing of the top surface or an increase of the Ni density. A density increase of at least 30% is needed to explain the measured reflectivity curve. This, however, is rather unlikely to happen[9]. Therefore, the first part of the curve is explained by a reduced surface roughness to less than 0.2 nm. The extreme (high) last oscillation can be explained by interface

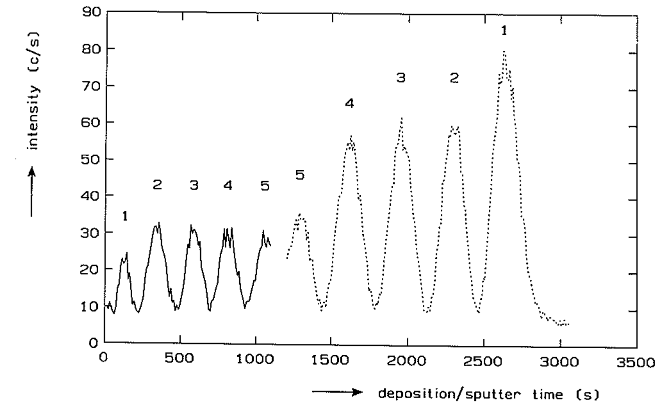

Figure 1. The reflectivity of N-K_α X-rays ($\lambda = 3.16$ nm, $\theta = 35°$) as a function of the Ni layer thickness, during growth (solid line) and during destructive sputtering using Ar^+ of 750 eV energy (dotted line). The maximum Ni thickness was 12.5 nm.

effects of the remaining Ni layer with the substrate (intermixing) or to surface relaxation effects at the Ni layer. This results in an increase in the Ni density[10] at the surface and determines the reflectivity when the layer thickness decreases.

The solid line of Figure 2 shows the X-ray reflectivity during growth of a Ni–C multilayer consisting of four periods (of 2.7 nm) with a thick Ni layer (12.5 nm) on top. Simulation revealed an increasing surface roughness during deposition from 0.4 to 0.8 nm. Agreement between the layer thicknesses derived from the X-ray reflectivity maxima and the quartz crystal was obtained, indicating that no density changes occurred within the monitor accuracy. Due to heat absorption, the film thickness of carbon could not be measured accurately by a quartz crystal monitor. The dotted line represents the X-ray reflectivity of the stack during sputtering of the film using Ar^+ ions of 750 eV (ion current = 1.5×10^{-5} A cm^{-2}). The reflectivity curve obtained during sputtering of the first 12.5 nm Ni can be simulated when a decrease of the surface roughness of more than 0.4 nm is assumed. The reflection pattern obtained during sputtering of the multilayer below the Ni film would suggest an increasing surface roughness or might be explained by interface roughening or mixing. Using the TRIM86 computer code to simulate penetration depths and recoil energies for Ar^+ ions of 750 eV impinging on a multilayer consisting of 1.3 nm Ni, 1.3 nm C and 17.4 nm Ni, at perpendicular incidence indicated firstly that intermixing of the first interface should be expected at a Ni thickness of about 1 nm, and secondly that the influence on the second interface is considerably less.

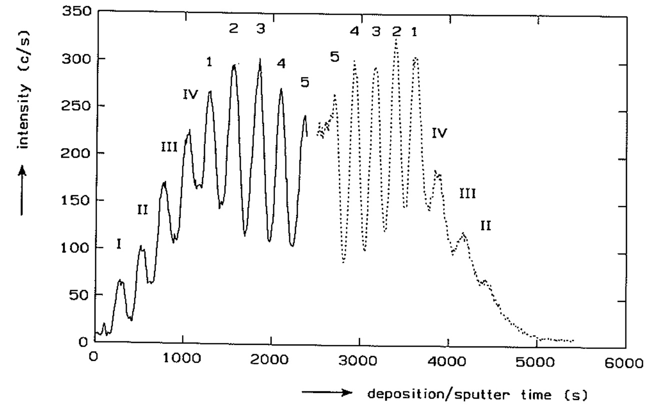

Figure 2. Solid line: The reflectivity of N-K_α X-rays ($\lambda = 3.16$ nm, $\theta = 35°$) for a Ni–C multilayer of four periods, having a periodicity of 2.7 nm and a thick Ni layer of 12.5 nm on top. Dotted line: the reflectivity during sputtering of the stack using Ar^+ (750 eV).

Figure 3 shows the X-ray reflectivity during growth (solid line) of a 10.0 nm nickel film, followed by sputtering (dashed line) with H^+ ions of 750 eV energy (ion emission is 1×10^{-5} A cm^{-2}). During growth the roughness increases from 0.4 nm of the Si(111) substrate to 0.7 nm for the 10.0 nm thick Ni film surface. The sputter rate calculated from the distances between the interference maxima is $\approx 1.5 \times 10^{-2}$ Ni atoms ion^{-1}, which is in agreement with measurements of Bohdansky *et al*[11]. The increased reflectivity fits a roughness decrease of about 0.6 nm.

Figure 4 shows the reflectivity of X-rays during the growth (solid line) of a multilayer of three periods with a thick Ni layer of 12.5 nm on top, followed by sputtering (dashed line) with H^+ ions of 750 eV (ion emission is 1×10^{-5} A cm^{-2}). During sputtering a dramatic decrease of the X-ray reflectivity can be observed. This decrease could only be understood assuming a complete destruction of the multilayer structure underneath the thick Ni film, by intermixing of the two materials.

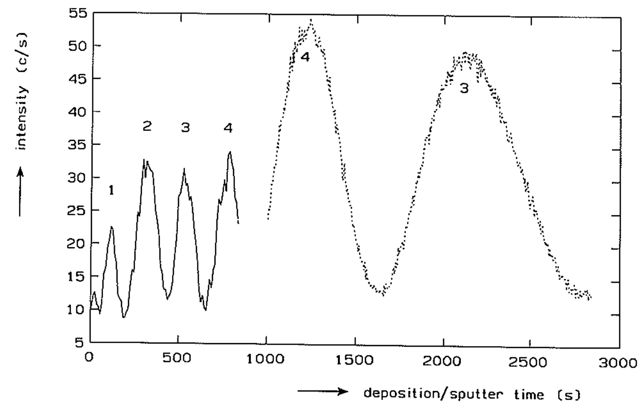

Figure 3. As Figure 1, now the total Ni thickness is 10.0 nm and sputtering has been done with H^+ of 750 eV.

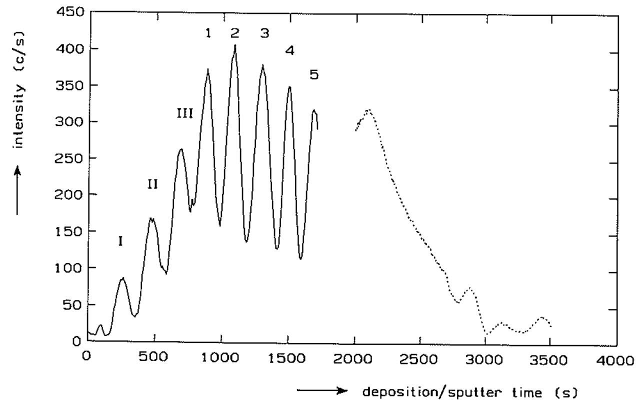

Figure 4. As Figure 2, now the Ni–C multilayer consists of three periods and sputtering has been done with H^+ of 750 eV.

The effect of H^+ ion bombardment has been simulated using TRIM86 with a stack consisting of three layers: 10.0 nm Ni, 1.3 nm C and 8.7 nm Ni. The penetration depth of H^+ ions turned out to be sufficiently large to cause intermixing of the Ni–C and C–Ni interfaces in a multilayer underneath a Ni film of 10.0 nm thick.

We explain the decreasing roughness of the surface of the top layer as follows. In the energy range used in our experiments, particles are removed from the surface by recoil effects. It is clear that the particles with the lowest binding energy, with the fewest nearest neighbours, will be removed first. Therefore particles on top of islands will be removed first, reducing roughness.

Conclusions

We conclude that depth resolution problems, when a combination of a surface sensitive technique and destructive sputtering is used, mainly arise from mixing of materials at the deeper interfaces and not from an increasing roughness of the top surface. Destructive sputtering with Ar^+ or H^+ both resulted in smoothing of the top interface. Intermixing occurred at interfaces below the first layer. This effect turned out to be more dramatic for H^+ than for Ar^+, due to a larger penetration depth for light ions.

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