

High resolution Rutherford backscattering spectroscopy studies on Mo/Si multilayers

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Abstract

High resolution Rutherford backscattering spectroscopy with an electrostatic analysis of the ion energy is applied to Mo/Si multilayers with a period of 7 nm. The multilayers have been produced for X-ray optical purposes by electron beam evaporation in ultrahigh vacuum at three different temperatures during deposition: 30, 150 and 200 °C. In the Rutherford backscattering spectra the layer structure is resolved in all three cases. The multilayers deposited at 150 and 200 °C show interlayers of mixed Mo and Si of different thicknesses on the two sides of a Mo layer. The most distinct layer structure is found for the 150 °C sample, whereas the spectra for the 30 °C sample indicate a larger interfacial roughness and those for the 200 °C sample larger interfacial layers of mixed Mo and Si than for the 150 °C sample. On baking the multilayers to temperatures higher than 400 °C, interdiffusion of Mo and Si is observed. The multilayers deposited at 150 and 200 °C are destroyed after baking to 600 °C, whereas the multilayer deposited at 30 °C has already been destroyed after baking to 500 °C. Up to a baking temperature of 600 °C neither losses of material from the stack nor accumulation of Mo or Si at the surface or the interface between the stack and substrate are observed.

1. Introduction

The Mo/Si multilayers, which are examined in this work by high resolution Rutherford backscattering spectroscopy (RBS), are designed as optical components in the soft X-ray range. They have been fabricated by electron beam evaporation in ultrahigh vacuum [1, 2]. Three different temperatures have been applied to the substrates during deposition (*in situ*): 30, 150 and 200 °C. The highest reflectivities (larger than 50%) are found for the multilayers deposited at 150 °C [2]. The thermal stability of the multilayers is of great importance in their application as X-ray mirrors. The multilayers fabricated at 150 and 200 °C are found to have a stable X-ray reflectivity even after baking them to 500 °C, while the reflectivity of the 30 °C sample is drastically reduced to negligible values at this baking temperature. After baking to higher temperatures this drastic reduction in X-ray reflectivity is observed also for the samples produced at 150 and 200 °C [3].

In this work, RBS is used to obtain the mass- and depth-resolved composition of the layer systems after deposition and after different baking steps. Up to now, RBS on multilayers with a period of less than 10 nm has been used only to determine the atomic ratio of the multilayer materials integrated over the whole stack (see for example refs. 4 and 5), because the at-

tainable depth resolution of a standard RBS system with semiconductor detector (depth resolution, greater than 10 nm) is not sufficient for resolving single layers. Therefore the measurements were carried out here by means of an electrostatic analyser, which provides sufficient depth resolution for a detailed study of the multilayer microstructure.

2. Experimental set-up

The experimental set-up is shown in Fig. 1. All measurements were carried out using a He⁺ beam provided by a 350 keV ion accelerator. The accelerator yields a current density of up to 80 nA mm⁻² on the target. We used a rectangular beam spot on the sample with a 1.8 mm horizontal size and a 0.9 mm vertical size. The targets (size, 14 mm × 28 mm) were mounted on a high precision manipulator, which allows three rotations and two translations lateral to the beam. The ion energy was determined by use of a toroidal electrostatic analyser. The analyser accepts an angular range of 25° in the scattering plane simultaneously. The energy resolution $\Delta E/E$ of the whole system (ion beam and analyser) is found to be 5×10^{-3} ; the angular resolution is better than $\pm 0.15^\circ$. The targets can be changed without breaking the vacuum by means of a target transfer system.

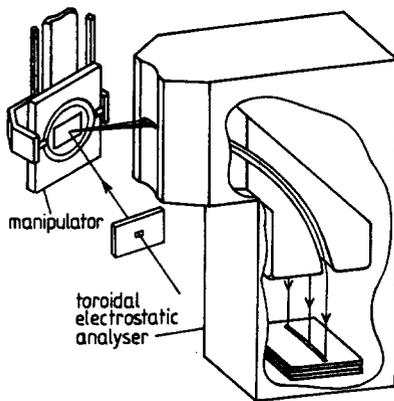


Fig. 1. Experimental set-up.

3. Results and discussion

In the lower part of Fig. 2 the backscattering spectra obtained with a 350 keV He^+ beam are shown for a Mo/Si multilayer consisting of 12 bilayers with a period thickness d of 7 nm which was deposited at a temperature of 150 °C. The spectra are measured in energy steps of 1 keV. After an ion dose of approximately $40 \mu\text{C mm}^{-2}$, ion-beam-induced mixing of the layer system became detectable; thus it was necessary to use a new beam spot on the target. By applying this ion dose, approximately 15–30 energy points could be measured in practice. No dependence of the backscattering yield from the orientation of the target due to ion channelling was observed; therefore the Si and Mo layers are amorphous or polycrystalline. Two different scattering geometries have been chosen: geometry a with normal ion incidence and an exit angle of 32° provides good mass resolution; geometry b with 30° ion incidence with respect to the surface normal and an exit angle of 70° results in a good depth resolution. For geometry a the signals of Mo (between 215 and 300 keV in Fig. 2(a)) and Si (below 210 keV in Fig. 2(a)) are separated, and both signals turn out to exhibit a number of oscillations which are due to backscattering from the individual layers in the stack. Since $\Delta E/E$ is constant over the energy, the analyser measures $E \times \text{intensity}$. This tends to compensate the increase in the Rutherford backscattering spectrum with decreasing energy for a constant material concentration in the target. Thus the spectrum measured directly with the analyser is in this case a good estimate of the distribution of the materials with depth. Because of the *in-situ* deposition technique the Si concentration remains virtually constant with increasing deposited thickness of the stack, while the Mo concentration decreases slightly. The energy resolution in the geometry a is limited owing to the isotope distribution of Mo, which results in an energy spread of about 4 keV, and owing to energy straggling, which

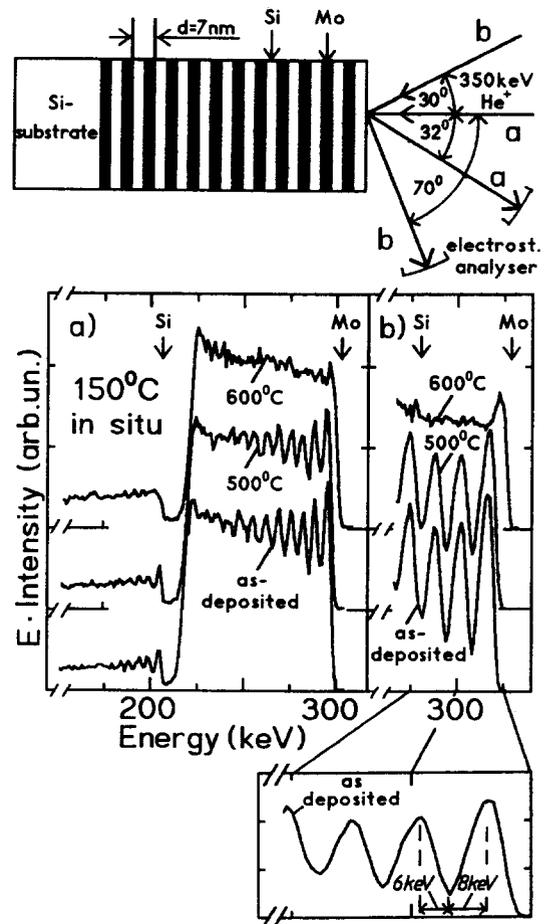


Fig. 2. Backscattering spectra for the multilayer fabricated at 150 °C with and without baking at temperatures of 500 and 600 °C. Two different geometries have been chosen: geometry a with normal ion incidence and an exit angle of 32° provides good mass resolution; geometry b with 30° ion incidence and an exit angle of 70° provides good depth resolution. The inset shows the spectrum for the non-baked sample with an expanded energy scale. The arrows in the middle part indicate the energies expected for Mo and Si for backscattering from the surface.

contributes an amount of approximately 2 keV to the energy spread at the first minimum in the Mo signal and increases with increasing path length in the sample. This results in the damping of the Mo and Si oscillations with decreasing energy in Fig. 2(a).

In geometry b the enhanced depth resolution allows a more detailed analysis of the formation of the Mo–Si interfaces. The minima in the lowest spectrum in Fig. 2(b) (the first three peaks of the spectrum are exclusively due to backscattering from Mo) are much deeper than those in Fig. 2(a), and the shape of the three peaks is asymmetric, which is clearly obvious in the inset with the expanded energy scale. The slope of the right-hand (top) side of the Mo peaks is much higher than the slope of the left-hand (bottom, with respect to the substrate) side. This is because layers of a Mo–Si

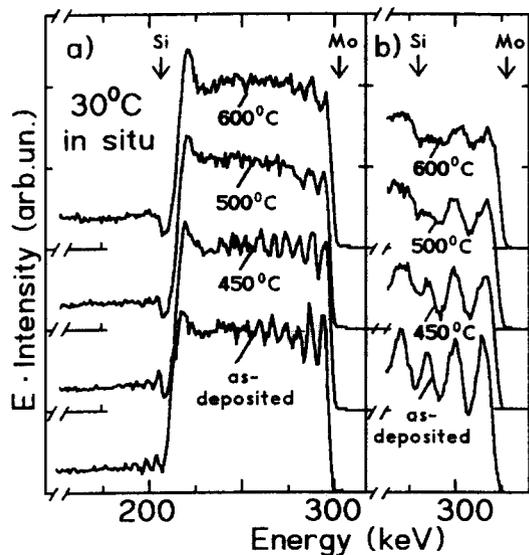


Fig. 3. As for Fig. 2 but for a multilayer deposited at 30 °C and baking temperatures between 450 and 600 °C.

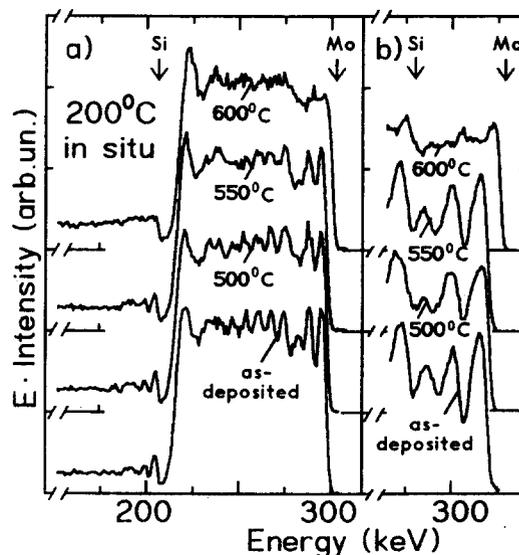


Fig. 4. As for Fig. 2 and Fig. 3 but for a multilayer deposited at 200 °C and baking to temperatures between 500 and 600 °C.

intermixed phase exist at the interfaces between Mo and Si and the thickness of these layers is considerably larger at the Mo-on-Si interface than at the Si-on-Mo interface [4, 5]*.

The multilayer was baked for 20 min at 500 and 600 °C in vacuum in order to observe the changes in the material distribution. The backscattering spectra obtained after baking are displayed in the middle and upper parts of Fig. 2. Baking at 500 °C has hardly any effect on the material composition in the stack. Only a slight damping of the oscillations in the spectra is observed. After heating to 600 °C both the oscillations in the energy distribution of the Mo and the Si part of the spectra have vanished. A shift to higher energies is observed for the high energy edge of the Mo part, while the intensity at the high energy edge of the Si part is reduced. These two facts are compatible with an interdiffusion of the Mo and Si layers. No redistribution of material over the whole stack is observed. The total intensity of backscattered particles in Fig. 2(a) does not change for the Mo part nor for the Si part, indicating that no material is lost during the baking procedure.

Corresponding studies have also been performed for the other two multilayers which were deposited at temperatures of 30 and 200 °C. The spectra for these samples are displayed in the lower parts of Fig. 3 and Fig. 4 respectively. Again a number of oscillations are observed in the spectra because of the distribution of

Mo and Si in the multilayer stack, but the oscillations are less pronounced than those for deposition at 150 °C (Fig. 2). This can be due either to interfacial roughness or to interlayers of mixed Mo and Si at the interfaces. Comparing the results for geometry b in Fig. 3 and Fig. 4 we note again (as for deposition at 150 °C) an asymmetric shape of the Mo peaks for deposition at 200 °C (Fig. 4(b), lowest spectrum), while this is not observed for deposition at 30 °C (Fig. 3(b), lowest spectrum). This is compatible with the findings in ref. 2 that the samples deposited at 30 °C exhibit a considerably larger interfacial roughness than do the 150 °C samples, while deposition at 200 °C results in sharper interfaces but with thicker interfacial layers of a Mo-Si mixture [3], which are obviously (as in the case of deposition at 150 °C) considerably broader at the Mo-on-Si interface than at the Si-on-Mo interface. Irregularities in the oscillations of the spectra, which are due to thickness errors in the stack, are also larger for deposition at 30 and 200 °C than for deposition at 150 °C. From the spectra in Fig. 4 we note for example that the thickness of the third Mo layer from the top of the stack is considerably smaller than that of the other Mo layers in the stack.

The multilayers deposited at 30 and 200 °C were also heated in order to test their thermal stability and to observe changes in the material distribution in the stack due to the baking procedure. The 30 °C sample was baked at 450, 500 and 600 °C and yields the Rutherford backscattering spectra in the upper part of Fig. 3. Corresponding spectra for baking at 500, 550 and 600 °C for the 200 °C sample are displayed in Fig. 4. In Fig. 3(a), damping of the oscillations in the spectra due

*Transmission electron microscopy studies with our samples (not yet published) have also shown thicker interfacial layers at the Mo-on-Si interface as in refs. 4 and 5.

to interdiffusion is already observed at 450 °C. At 500 °C the oscillations have disappeared almost completely with exception of the first two Mo layers from the top of the stack. For the 200 °C stack in Fig. 4(a), oscillations in the spectra are even observed after a baking procedure to 550 °C. In this case the oscillations in the spectra vanish almost completely after baking at 600 °C. When comparing the three spectra for geometry b (in Figs. 2–4), after baking to 600 °C, it can be noted that Mo oscillations still exist for the first layer (150 °C sample (Fig. 2)) or for the first two layers (Fig. 3 and Fig. 4) of the stack. This is probably because the silicon spacer is completely consumed after baking at 600 °C. In addition, a contribution due to backscattering from oxygen is contained in the spectra, which corresponds to the surface region of the stack and is therefore probably due to a silicon oxide layer at the surface. Since the silicon oxide does not interact with Mo at temperatures below 600 °C, this can additionally be responsible for the fact that the Mo oscillations near the surface region have not completely vanished.

A quantitative analysis of the spectra yields the result that for all three samples the integrated backscattering intensity for both the Mo and the Si contribution does not change during the baking procedures. Neither a loss of material nor a supply of material from the substrate to the stack is therefore found. This conclusion is also supported by the fact that the low energy side of the Mo contribution to the spectra shows a steep increase at approximately 215 keV corresponding to the interface of the substrate (Si(111) with a thin silicon oxide

layer) with the first Mo layer in all spectra and that the steepness and the energy position* do not change during the baking procedure. Since X-ray reflectivity data [3] show a considerable change in d spacing after baking, it can be concluded that this change in d spacing is exclusively due to interdiffusion processes within the stack with a corresponding increase in average density.

Acknowledgments

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*The slight shift to higher energies for baking to 600 °C can be explained by intermixing of Mo and Si within the stack.