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# CONCERNING THE PROBLEM OF HIGH DEPTH RESOLUTION USING ION SPUTTERING

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## ABSTRACT

The inhomogeneity of ion bombardment, the angular dependence of sputtering yields and the crystalline orientation of samples are the three main causes of the degradation of resolution with depth. It is possible to reduce these effects by bombarding at low energy (≈ 1 keV). A low-energy ion-bombardment device is described which has been adapted for use on our sputtered thermal-ion source mass spectrometer.

Key Words: Ion sputtering, mass spectrometry, depth resolution, sputtering yield.

## INTRODUCTION

The study of surfaces brings into play many competing analytical methods which are very often associated with ion etching techniques to explore the sample composition at depths varying from nanometers to micrometers. The ion etching technique has really become an essential tool in surface science. Depending upon the technique to be used, ion erosion conditions are very different : the sputtered area may vary from a few 100 $\mu m^2$ in Secondary Ion Mass Spectrometry (SIMS) or Auger Spectroscopy to about 1 $cm^2$ in Electron Spectroscopy for Chemical Analysis (ESCA) ; the ion energy from ≈ 1 keV to 10 keV and the beam density from a few hundred $\mu A/mm^2$ to some tens of $\mu A/mm^2$. Therefore, it is not surprising that under these circumstances the comparison of depth profiles is extremely difficult. Often, concentration profiles are plotted as a function of the sputter time with no reference to the beam density over the image field defined by the optical system of the method or to the erosion rate, which renders the depth calibration difficult to determine.

However, independently of the problem of calibration - that can be solved by using the mass thickness concept (Blaise, 1985) - the major problem of ion erosion is the depth resolution. This may vary to a large extent depending on the erosion procedure used, in such a way that it is not easy to understand the physical significance of a profile without information about the resolution for the conditions in which it was obtained.

The causes of resolution degradation with depth are multiple but there is now a convergence to express the loss of resolution by an expression of this form (Hofmann,1980, Mathieu et al., 1976, Laty et al., 1979) :

$$\Delta z \sim \alpha + \beta z^{\gamma} \quad (1)$$

where z is the depth, $\Delta z$ the dispersion of depth characterized by the constants $\alpha$ and $\beta$ and $\gamma$ an exponent varying from 0 to 1. There are many papers discussing the $\gamma$ value and its physical meaning.

The causes of the loss of resolution can be classified in three categories :

i) Sputtering conditions : the inhomogeneity of the ion beam, the nature and energy of ions, the incidence angle to the surface.

ii) Sample texture : multiphase texture producing local variations of the sputtering yield, polycrystalline structure leading to variation of the erosion velocity from one grain to another (Blaise, 1978), initial roughness inducing the formation of cones (Carter et al., 1983, Limoge, 1984).

iii) Collisional processes : atomic mixing and formation of an altered surface layer (Littmark and Hofer, 1980), collapse of defects in dislocation loops producing a local variation of the sputtering yield (Hermanne, 1973) and enhanced diffusion due to the formation of atomic defects (Seran and Limoge, 1981). Some of these causes, included in the term $\alpha$ of expression (1), produce a loss of resolution nearly independent of the eroded thickness - atomic mixing, altered layer - but most others contribute to an increase in the loss of resolution with depth. This is why it is so important to look for a procedure of erosion which minimizes the variation in resolution with depth.

## THE MAJOR CAUSES OF THE LOSS OF RESOLUTION WITH DEPTH

There are three major causes of loss of resolution among those mentioned above :

i) the inhomogeneity of ion bombardment.
ii) the angular dependence $S(\theta)$ of the sputtering yield, which is responsible for the formation of cones (Carter et al., 1983).
iii) the crystal orientation dependence of the sputtering yield which makes a polycrystalline surface look like a Louis XIV pavement after an extended period of erosion (Bernheim, 1972).

### *Inhomogeneity of bombardment.*

There are two procedures to obtain a beam of homogeneous density on the analyzed area, viz. a defocusing of the beam or a scanning of the beam across the surface.

The defocusing technique is simple but not adapted for all situations. Let us suppose a gaussian beam of width $2\sigma$. A simple calculation of the $\sigma$ value required to obtain uniform density over an area of a diameter 2 x a perpendicular to the beam axis, with a precision of p leads to :

$$\sigma = \frac{a}{\sqrt{2|\ell n\ (1-p)|}} \qquad (2)$$

If p = 0.01 is required one obtains $\sigma \approx 7$ a. If the image field is relatively small, i.e. a is $\approx 100\ \mu m$, as in the Ion Microanalyser, a spot of 2 mm in diameter is sufficient to obtain a uniform erosion. This is quite reasonable for a beam whose intensity is several $\mu A$. If uniformity is required over a large area, typically 1 $mm^2$, the surface of the beam must be $\sim$ 150 $mm^2$ which results in a bombardment with a very low density, that is at a low erosion rate. Erosion under these conditions is generally not recommended because of the influence of the residual atmosphere on the surface during erosion.

In this procedure, the inhomogeneity of the beam imposes a minimum precision $\Delta z/z$ = constant which corresponds to $\gamma = 1$ in expression (1).

A scanning of the beam on the surface is better adapted to most situations. In a raster mode it allows a visualisation of the surface by secondary electron emission but it may induce under certain conditions a surface structure which limits the resolution as in the defocusing mode (Degrève et al., 1979). It seems that the best scanning mode consists in fact of applying on each pair of deflecting plates a symmetric voltage of two different frequencies : a low frequency in the x-direction and a high frequency in the y-direction.

Let us consider a squared area of the sample to be sputtered by a beam of diameter $2\sigma$. To simplify the problem, we suppose the beam is moved step by step by a constant increment of voltage $\Delta V$. The number of steps in both directions is $n = V_m/\Delta V$ where $V_m$ is the amplitude of the two periodic voltages. The number of elemental areas covered by the spot every second is $2n \frac{f_y}{f_x}$, that is, on the average, $2 \frac{f_y}{n}$ impacts per second at the same point. Assuming the displacement is at random, one obtains a fluctuation of intensity at any point of $p \% = \sqrt{\frac{n}{2f_y}}$ for one second of bombardment. After a time t of bombardment the fluctuation is reduced to $p \%/\sqrt{t}$. We see immediately the advantage of the method : the fluctuation of depth decreases as the erosion is progressing. Therefore, the homogeneity of erosion is not a limiting factor of resolution.

The only precautions to be taken are to eliminate the edge of the bombarded area, from the image field, to a width of the order of the beam diameter (because the dwell time is not the same at the edge as in the center of the bombarded area) and to choose the two frequencies such as to prevent a Lissajous configuration of low index.

Let us illustrate this procedure with the example of the Sputtered Thermal Ion Mass Spectrometry method (STIMS) (Blaise, 1985). The beam diameter is $2\sigma \sim 0.5$ mm, $n \sim 6$, $f_y \sim 3000$ Hz and $f_x \sim 300$ Hz. This gives a fluctuation of depth of 3 % in one second, that is a depth equivalent to about 3 % of a monolayer. This fluctuation has of course an influence on the precision of measurements. Fortunately as elements are simultaneously detected in this method there is a compensation of the fluctuations which improve the precision.

*The angular dependence of the sputtering yield.*

The sputtering yield $S(\theta)$ varies with the incidence angle $\theta$ to the normal to the surface. It increases from a minimum value $S_0$ at normal incidence to a maximum at an angle $\theta_c \approx 70°$ and then declines towards zero at grazing incidence (Carter et al., 1983). In the upward part of the curve (fig. 1) $\theta < \theta_c$, $S(\theta)$ is expressed as :

$$S(\theta) = S_0 (\cos \theta)^{-n}$$

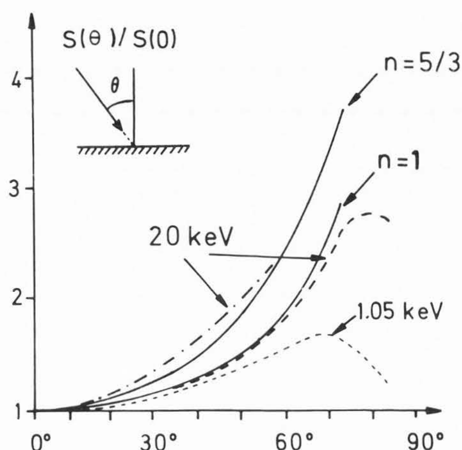


*Fig. 1*

*Dependence of the sputtering yield S(θ) with the incidence angle. For references, see Blaise (1978).*

Sigmund's theory predicts n = 5/3 (Oechsner, 1975). Depending on the ion energy, experimental values of n are situated on each side of n = 1. At high energy (20 kV) n > 1 is in good agreement with theory (for references see Blaise, 1978) whereas at low energy ($\approx$ 1 keV) the exponent n is < 1 (Oechsner, 1975).

This angular dependence of the sputtering yield is the cause of the formation of microstructures composed of cones, pyramids, edges (Carter et al., 1983). It has been shown that these protuberances develop only when the critical angle $\theta_c$ of maximum sputtering is present in the initial surface topography.

The sputtering of an unannealed microcrystalline vapor-deposited layer by ions of several keV energy is a typical example of the formation of cones. In figure 2, the density of cones obtained after sputtering of a 2,000 Å thick aluminium layer makes the surface look like the Black Forest in Germany. In such samples the height of cones increases with the depth of erosion without a hope of obtaining a final planar polishing of the surface. According to the interpretation, it is concluded that $\theta_c$ is present in the initial surface structure, probably at the grain boundaries.

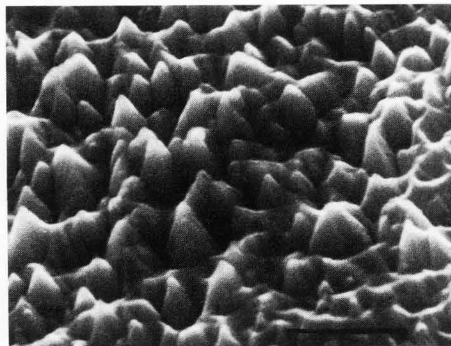

*Fig. 2*

*Formation of cones of a 2000 Å evaporated aluminium layer bombarded by $Ar^+$ ions of 5 keV (magnification × 20,000 ; bar = 1 µm). The depth of erosion exceeds the layer thickness.*

The rate of erosion $v_\perp$ of a surface element in a perpendicular direction to the surface is :

$$v_\perp = \frac{N_p \, S(\theta) \cos \theta}{N} \qquad (3)$$

where $N_p$ is the primary beam density and N the atomic density of the solid.

The best sputtering conditions are those producing a uniform rate of regression $v_\perp$ of each surface element, whatever its angle of incidence to the ion beam. This implies the condition $S(\theta) \cos \theta$ = constant. Therefore a compensation between $S(\theta)$ and $\cos \theta$ must occur to obtain a good erosion. This compensation is roughly achieved up to the critical angle $\theta_c$ but, beyond this value no compensation is possible at all. This explains the formation of cones if incident angles $\theta > \theta_c$ are present at the surface.

The formation of cones leads to a rapid degradation of depth resolution. Thus it is essential to establish sputtering conditions which would reduce or eventually prevent the development of protuberances of any kind. It is expected, for example, that the light dependence of $S(\theta)$ with $\theta$ at low bombarding energy is favorable to obtain a better resolution.

*Crystalline dependence of the sputtering yield.*

When a solid remains crystalline under ion bombardment as for metals, an influence of the atomic arrangement on the sputtering yield is observed (Bernheim, 1972, Blaise, 1978). The lattice structure has an effect on the penetration of bombarding ions and ejection of secondary particles.

A dependence of the sputtering yield with the crystallographic index planes of copper single crystals bombarded at normal incidence is shown in fig. 3 (Southern et al., 1963). At 5 keV energy there is a factor of 4 between the two extreme

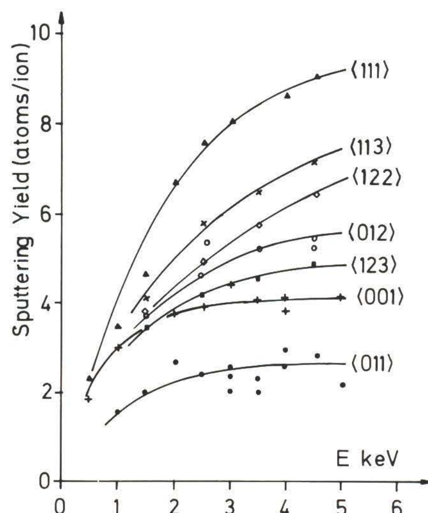


*Fig. 3*

*The energy dependence of the sputtering yields of copper single crystals bombarded at normal incidence by $Ar^+$ (Southern et al., 1963).*

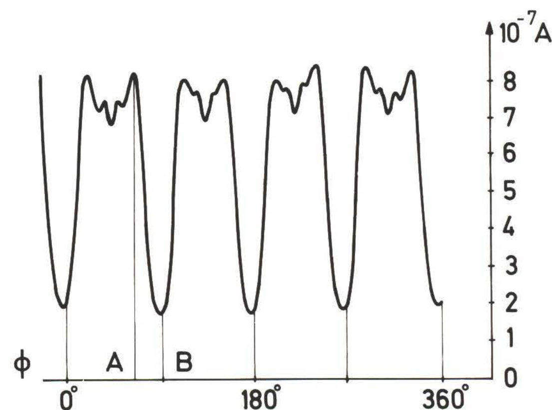


*Fig. 4*

*Bombardment of a (100) aluminium single crystal by $Ar^+$ of 6 keV at an incidence θ = 45°. Variation of the secondary ion emission $Al^+$ signal as a function of the azimuthal angle. A minimum (B) is obtained when the beam is directed along the <110> row (transparent direction). When the bombardment is in an opaque direction (A) the emission is maximum.*

yields corresponding to (011) and (111) planes. When the bombarding energy is decreased, the dispersion of yields is considerably reduced.

The effect of the lattice structure on the penetration of the primary beam has been investigated by Bernheim (1973) in an impressive experiment performed on a (100) Al crystal bombarded at constant incidence angle of 45° by $Ar^+$ ions of 6 keV. In this experiment, secondary ions whose intensity is proportional to the sputtering yield were recorded as a function of the azimuthal angle. This angle is varied by rotating the sample around the normal to the surface. A strong variation of S with the rotating angle is observed, with deep minima when the beam is directed along low index crystallographic directions <110> (fig. 4). These variations of S are due to the transparency of the crystal relative to the beam incidence. The effect can be understood in a simple hard-sphere collision model (Odintsov, 1963). In such a model each atom is represented by a sphere whose radius is a few tenths of Angströms. A collision occurs with a lattice atom when the trajectory of the primary particle intercepts a sphere. When the bombardment is directed along a <110> row, atoms of the top layer mask atoms of underlying layers. This direction of bombardment is referred to as transparent direction. It corresponds to a low sputtering yield because only atoms of the top layer are hit by a primary ion and initiate microcascades sufficiently close to the surface to produce atomic ejection in the vacuum. Ions that penetrate deeply into the crystal are inefficient for sputtering. Now if the bombardment is in a direction referred to as opaque, where there is no masking between hard spheres of atoms located in the first few layers, the collision probability close to the surface is increased and, consequently, the sputtering yield.

A quantitative study of transparency effects (Laurent, 1973) has shown that the sputtering yield is proportional to the collision cross section of a primary atom in the first few atomic layers. Therefore it can be concluded that the sputtering of crystalline solids results essentially from microcascades initiated by the collision of the primary particles with atoms of the first atomic layers , most of the ejected atoms coming from the top layer. With ions of a few keV energy - typically 5 to 10 - collisions in the first four atomic layers must be involved in order to interpret the crystalline dependence of the sputtering yield. This corresponds approximately to the extreme variations of S within a factor of 4.

Effects of crystalline texture on sputtering may seriously complicate the problem of depth analysis since the grains of a polycrystalline sample will be eroded differently according to their respective orientation to the beam. Bombarding at ∿ 5 keV the variation of the erosion velocity from one grain to another will be about a factor of 2 on an average. This means that the depth resolution which is expressed as $\Delta z/z$ cannot be better than about 25 %. This is effectively what we observe when polycrystalline samples are bombarded.

Several procedures have been proposed for eliminating transparency effects (Bernheim, 1973), most popular one consists of flooding the sample surface with reactive gas, as oxygen, in order to produce an amorphisation of the surface under bombardment (Blaise, 1978). This procedure was successful in a number of cases but this is not general. Furthermore, in some applications oxidation or any other treatment of the surface must

be excluded. This is why it is so important to use sputtering conditions where transparency effects are attenuated. From the results reported in fig. 3, the dispersion of single-crystal sputtering-yields decreases when the ion energy diminishes. For example, at 1 keV, sputtering coefficients are confined within a factor of 2 whereas this factor is about 4 at 5 keV. This feature is confirmed by the measurement of the contrast of curves like the one presented in fig. 4, for crystals of any orientation. If the contrast is expressed by the ratio of the sputtering yield measured in a given orientation of the bombardment to the average yield, one gets 0.34 at 6.2 keV, 0.25 at 4.8 keV and 0.16 at 2.4 keV (Bernheim, 1973). Finally, it is to be expected that by reducing the bombarding energy, the crystalline dependence of S is considerably attenuated.

The conclusion of these two last discussions demonstrates the necessity to use a low energy sputtering device to obtain depth analysis under the best conditions. Such a device has been attached to our Sputtered Thermal Ion Source Mass-Spectrometer (Blaise, 1985).

## CHARACTERISTICS OF SPUTTERING AT LOW ENERGY

### *Low energy and high flux ion gun.*

Good sputtering conditions first require the bombarding of the sample area to be analyzed with a sufficiently high ion density, typically in the range of 0.1 to 1 $\mu A/mm^2$. In our technique (Blaise, 1978) the beam must be scanned over about 8 $mm^2$ to get a uniform erosion on the image field of the order of 1 $mm^2$. Therefore an intensity of a few $\mu A$ is necessary.

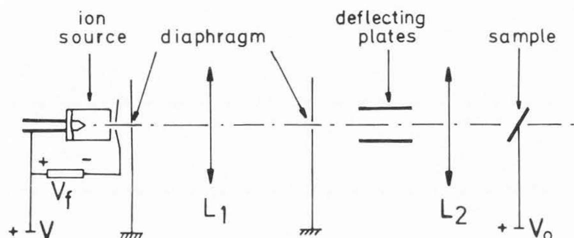


*Fig. 5*

*Schematic view of the ion gun attached to the sputtered Thermal Ion Source Mass Spectrometer (Blaise, 1985).*

In the device represented in figure 5, ions are extracted at a fixed voltage $V_f$ conveniently adapted, from a conventional filament source. The source plus the extracting electrode are brought to a positive voltage V. The emitted ions have an energy eV independent of the extracting voltage $V_f$ when they pass through the diaphragm 0 at ground potential. Two lenses $L_1$, $L_2$ associated with two pairs of deflecting plates bring the beam to the sample. Differential pumping provides a good vacuum ($\approx 10^{-6}$ torr) in the sample chamber. The intensity delivered to the sample versus the accelerating voltage is shown in figure 6.

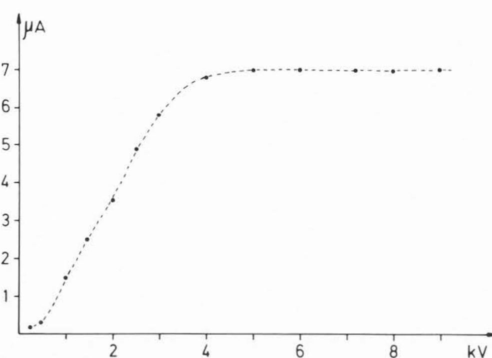


*Fig. 6*

*Intensity delivered by the ion gun to the sample as a function of the accelerating voltage V.*

In the mass spectrometer, the sample is at a positive voltage $V_0$ = 4 kV, therefore ions hit the target with an energy $e(V - V_0)$. It is seen in fig. 6 that the ion intensity remains constant over an extracting voltage of 4 kV. Therefore it is possible to bombard the sample with a constant intensity, from the threshold sputtering energy up to several keV. However the threshold sputtering energy is not measurable with accuracy because there is no energy filtering on the ion beam.

### *Dependence of S on ion energy.*

Absolute measurements of S, which are extremely difficult to obtain (Andersen and Bay, 1983), are not of a real interest for analytical purposes because just a small part of the sputtered matter is collected in the mass spectrometer. Therefore from a practical point of view, it is sufficient to study the energy dependence of the collected sputtered atoms.

In our technique, the sample is bombarded at an incident angle of 45° and sputtered atoms are collected symmetrically in the heated cell, through the small opening 0 (fig. 7). Let $\Delta N$ be the number of atoms collected per second and $\Delta n^+$ the number of atoms detected in the mass spectrometer.

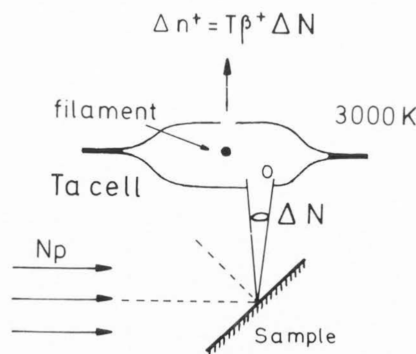


*Fig. 7*

*Schematic view of the collection of sputter matter in the Thermal Ion Source Mass Spectrometer.*

One gets :

$$\Delta n^+ = T\ \beta^+\ \Delta N \qquad (4)$$

where $\beta^+$ is the ionization coefficient and T the transmission factor. We have plotted in fig. 8 the variations of $\Delta n^+$ versus the energy of argon ions, for three typical metals : a light one (Al), a medium one (Cu) and a heavy metal (U). Above the threshold energy which is in the range of 25-50 eV the three curves have in common a rapid increase of $\Delta N$ with energy up to $\approx$ 1 keV. Then, above 1 keV, there is a diversification of behaviour : a rather large increase of $\Delta N$ for the light metal, a slower increase for copper and a plateau for the heavier metal.

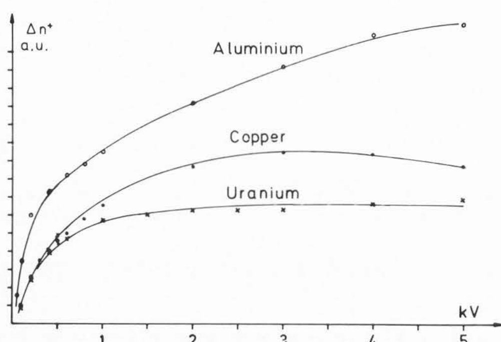


*Fig. 8*

*The energy dependence of the signal $\Delta n^+$ detected for three metals (aluminium, copper and uranium) bombarded by $Ar^+$. Intensities are in arbitrary units.*

These experiments show that it is possible to sputter any sample with an energy as low as 1000 eV or so, with a reduction in the yield that does not exceed a factor of 3 compared to the yield obtained at a bombarding energy of a few keV. Therefore bombardments at low energy appear extremely promising if an appreciable improvement in resolution is to be obtained.

## *Crystalline effects and cone formation.*

To determine the conditions in which crystalline effects and cone formation are minimized we have performed two types of experiments.

In the first one the $Cu^+$ signal coming from three copper samples composed of large crystals several hundreds of microns in diameter were recorded as a function of the bombarding energy. The results are presented in fig. 9. For the three samples the behaviour is the same up to $\approx$ 1 keV. Beyond this energy a dispersion in intensities is observed. The number of micro-crystals in the image field (2 to 5) is too small to allow averaging of the sputtering yield. So, if crystalline transparency plays a role in the sputtering process the measured intensities $\Delta n^+$ will not be the same due to the different grain orientation. This is what occurs above 1 keV, which shows that sputtering is appreciably affected by crystalline effects above this bombarding energy.

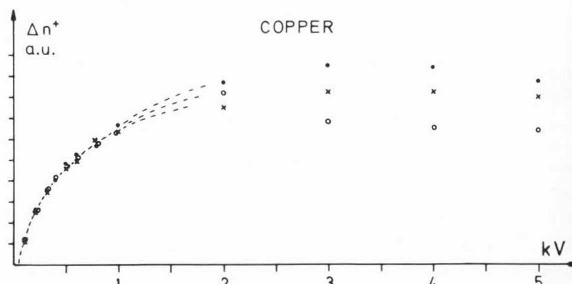


*Fig. 9*

*The energy dependence of the signal $Cu^+$ detected for three copper samples bombarded by $Ar^+$. Symbols • × and ∘ refer to measurements obtained from each sample. Intensities are in arbitrary units.*

The second experiment concerns the formation of cones. Several evaporated aluminium layers 2000 Å thick were bombarded at different energies by argon ions and the surface examined after bombardment in a scanning electron microscope. The topography obtained at 5 keV and 1 keV is shown in figures 2 and 10. There is a remarkable difference between the two structures. At high energy we have always observed the formation of cones whose height is comparable to the layer thickness. Below 2 keV energy a strong attenuation of cones is observed so that they completely disappear at 1 keV. The depth resolution is considerably improved in that case (Blaise, 1985).

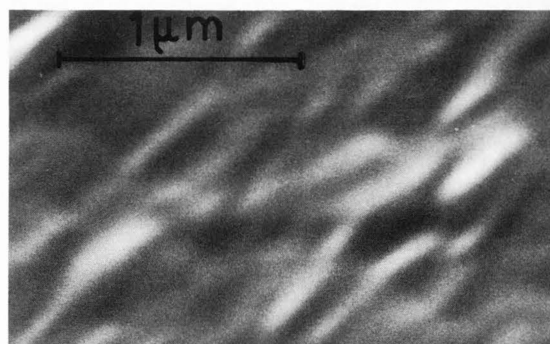


*Fig. 10 Surface topography of a 2000 Å evaporated layer after bombardment by $A_r^+$ of 1 keV (magnification, 50,000. Bar = 1 μm).*

## CONCLUSIONS

Most of the instruments which utilize sputtering are not designed with considerations to the problems specific to depth analysis. As a result, they do not work under the best conditions for resolution. Our work is an attempt to rationalize the problem of depth resolution on our instrument.

It is perfectly clear that low energy sputtering is a condition necessary to obtain a good resolution. But the problems posed are of two types : first, to obtain a sufficient density of bombardment and second to reconcile the energy to be used with the sputter yield. The results show

that the best conditions are for energies lying between 500 and 2000 eV. Perhaps a compromise of about one keV would be acceptable.

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## Discussion with Reviewers

R. Gijbels : What is the influence of the residual atmosphere (or oxygen flooding) for selecting appropriate beam parameters ?
Author : Oxygen flooding contributes to improving the depth resolution by reducing the crystalline dependence of the sputtering yield (see Bernheim, 1972). But, on the other hand, surface oxidation may also generate the formation of cones under ion bombardment. For the moment we have no rule for selecting the most appropriate depth analysis parameters for combining oxygen flooding, ion energy and ion beam incidence.

R. Gijbels : The author shows clearly the beneficial effects on depth resolution by using low-energy primary ions. How easy is it to implement such experimental conditions, in practice,on typical commercial SIMS instruments ?
Author : It is not easy to attach a low energy sputtering equipment on conventional commercial SIMS instruments. But it seems that an effort in that way is projected for the following generation.

W.B. Robinson : The ion count rate shown in figures 8 and 9 as a function of primary beam energy may indeed reflect changes in the sputter yield, as proposed, but it is also likely that these changes in ion intensity reflect changes in 1) the angle at which the primary ion beam strikes the sample,i.e.,changing the primary beam energy also changes the angle at which the ions strike the surface and 2) the ionization efficiency of the Thermal Ionization Mass Spectrometer.
Author : The objections raised by the reviewer are possibly those of a research worker familiar with SIMS. In our techniques there is no electric field in the sample chamber and the ionization efficiency is not dependent on sputtering conditions (see Ref. Blaise, 1985). As a consequence the angle (45°) at which the primary ion beam strikes the sample and the ionization efficiency are independent of primary energy. The ion count rate variations shown in fig. 8 and fig. 9 are really due to the variation of primary energy.

R. Gijbels : The angular dependence of the sputtering yield, for $\theta < \theta_c$ is given by $S(\theta) = S_0 (\cos \theta)^{-n}$ with experimental n values in agreement with theory, at least at high ion energy. Is there also a theory available for

$\theta > \theta_c$ with which experimental data (fig. 1) could be compared ?
Author : To my knowledge there is no theory available for $\theta > \theta_c$.

S. Hofmann : For incidence angles below $\theta_c$, a dependence $S(\theta) = S_o(\cos\theta)^{-n}$ with n = 1 should yield a minimum effect on sputtering induced roughness (see e.g. Seah and Lea, 1981). Could you please comment on that question and on the dependence of induced roughening with the ion incidence angle $\theta$ ?
Author : The problem discussed in detail by Seah and Lea (1981) is the uniform regression of a rough surface composed of a distribution of microplanes. As the local primary beam density varies as cos $\theta$ where $\theta$ is the local incidence angle, a compensation is achieved on each surface element if $S \sim (\cos\theta^{-1})$(see relation 3 in the text). In fact $S \sim (\cos\theta)^{-n}$ with n different from unity and varying from one element to another. Therefore no exact compensation is possible on a large range of angle $\theta$. Compensation is just possible for $\theta \sim 0$ which implies that bombardment is directed normally to the average surface and that the angle distribution of microplanes to the average plane surface does not exceed a few degrees. From this point of view the incidence angle of the primary beam plays an important role on the depth resolution : a better resolution is obtained for a bombardment aligned along the average surface normal.

Another problem is to prevent the formation of a microstructure (cones, pyramids..) superimposed to the initial roughness. Let us suppose that some surface elements are inclined at large angles $\theta \gtrsim \theta_c$ under the beam, as is, for example, the case at grain boundaries on a microcrystalline evaporated layer. At any incidence of the primary beam to the average surface plane, some surface elements will be inclined at those critical angles $\theta \gtrsim \theta_c$. Therefore in any case a microrelief composed of cones, pyramids.. may be generated. Thus I would say that the incidence angle is probably of little importance in that case. Another parameter such as energy, for example, must be involved to overcome the problem of the formation of a microstructure.

S. Hofmann : The measurements reported in fig. 9 is valid for secondary emission in a certain angle. Is this really representative for the integral mean sputtering yield which determines the erosion rate of the sample ?
Author : In a number of cases we have observed a good qualitative correlation between the emission in our collection angle and the measured erosion depth which is representative of the integral mean sputtering yield. Thus, from the results presented in fig. 9 we can estimate, for example, that the sputtering yield of the crystal whose measurements correspond to black dots is higher than the yield of the crystal whose measurements are represented by open dots. However, it is not possible to deduce integral mean sputtering yields with accuracy from measurements given in fig. 9.

A. Lodding : In your apparatus (fig. 7) you bombard at a fixed angle and collect the secondary particles through a fixed opening 0, in order to determine the dependence of S on the primary ion energy $E_p$. Is it not possible that also the ejection angles are dependent on $E_p$ ?
Author : The problem raised in this question is the dependence of the angular distribution on the primary energy. If we leave out primary energies close to the threshold sputtering energy and grazing incidence or emission angles, the shape of the angular distribution is not very dependent on primary energy (see Wehner and Rosenberg, 1960). Therefore, the curves presented in fig. 9 are qualitatively representative of the evolution of S with primary energy.

W.B. Robinson : The problem of depth resolution is not addressed in a straightforward manner and that is the major topic of the paper. It should be elementary to monitor the ion count rate of a thin film on a substrate (as it is sputtered away) as a function of primary ion energy and thus provide concrete evidence -by the use of eq.(1)- of improved depth resolution.
Author : This is a pertinent remark of common sense. First, an example of the effect of energy has been published in this review (Blaise, 1985). But this is a specific case among many others that could be found in the literature.

In writing this paper my purpose was not to show one or two more examples of this type but to deal with the problem of depth resolution in a more general way. Results reported in this paper are the first of a large investigation which is carried out to classify materials according to the ultimate depth resolution obtained. It is not an "elementary problem" to determine this resolution : special attention must be brought to the nature of the substrate, polishing, sample layer preparation...

R. Gijbels : What experimental conditions would be preferable for depth profiling shallow implants e.g. what angle $\theta$ for a given primary ion energy ? What would be the minimum layer thickness which can be detected by SIMS ?
Author : It is too early to answer the first part of this question in a rational way because no systematic study has been done up to now. I would say an incident angle between 45 and 60° and a primary energy as low as possible, compatible with the sputtering yield.

About the second part of the question there are many values published in the literature (see SIMS III, 1982) obtained for specific cases. In fact there is not a unique answer. The minimum layer thickness detectable may vary not only with the sample texture but also with the metallurgical properties of the material.

R. Levi-Setti : The transparency model of Odintsov is definitely obsolete, in view of Lindhard's channelling theory, and its application to the interpretation of the experimental data by Onderdelinden (1966, 1968). My feelings on the formation of cones and pyramids due to sputtering in crystalline and polycrystalline materials, is that channelling must play an important role (see e.g., the experiments by Francken and Onderdelinden, 1970). There is an energy dependence $\sim E^{-1/4}$ of the width of the channelling angles (axial channelling) and $E^{-1/3}$ (planar channelling), which probably plays a role in the observed disappearance of cone

formation at low energies. Furthermore, as the range of the incident ions becomes shorter and shorter, the "random" fraction of the beam will become dominant since there is not enough penetration in the crystal to establish a channelling regime.

Author : Thank you for this question which is of major importance. The concepts of transparency and channelling have some similarities : a low index crystallographic direction of channelling is also a direction of high transparency ; in the same way a high index crystallographic direction of dechannelling is a direction of low transparency.

Channelling which is associated with the stability of the particle trajectory along dense atomic rows, implies a deep penetration. On the contrary, transparency is related to shadow effects and can be reduced to the useful thickness of the crystal that is to a few atomic layers in the case of sputtering. I would say that the concept of channelling is a consequence of transparency. There is no doubt that channelling has an effect on the crystallographic contrast that you observe in your Scanning Ion Microscope (Levi-Setti et al. 1983 and La Marche et al. 1983) bombarding with high energy ions, as it also explains the angular dips indicated in fig. 4, although the bombarding energy is lower. But channelling cannot explain all the contrast, in particular the fine structure observed between two dips in fig. 4. To account for this structure we must introduce the concept of transparency. I don't think that the transparency model of Odintsov is definitely obsolete although its formulation is somewhat naive. As an example I will show the results of an experiment similar to those presented in fig. 4 but with a single crystal of any orientation. To account for the contrast observed in figure 11, it is just necessary to involve the transparency of the first four atomic layers relative to the beam orientation. Furthermore it is possible to interpret the evolution of the contrast with temperature (Laurent, 1973 ; Laurent, Slodzian, 1973).

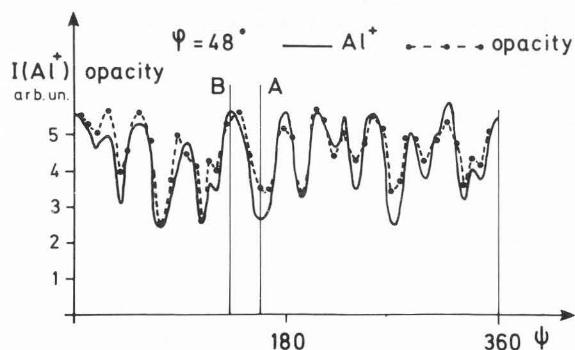


Fig. 11

Bombardment of an aluminium single crystal of any orientation by $A_p^+$ ions of 6 keV at an incidence θ = 48°. Full line : variation of the secondary ion emission $Al^+$ signal as a function of the azimuthal angle. Dotted line : variation of the transparency including the first four atomic layers calculated in a hard sphere collision model (Laurent, 1973).

I am convinced that the transparency model is well adapted to interpret the contrast in secondary ion emission, which is due to variation of sputtering yield. Now, about the role of channelling in the disappearance of cone formation at low energies, as you suggested it, there is no experimental evidence, to my knowledge, of any objection. The problem is still open.

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