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*Sputtering by Particle Bombardment,
Experiments and Computer Calculations
from Threshold to MeV Energies*

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Introduction and Overview

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Abstract. The physics processes causing sputtering, i.e. the removal of atoms from the surface of solids or liquids at bombardment with particles having energies from the eV to the MeV range are today mostly understood. The erosion yields calculated with computer codes for different incident parameters agree reasonably well with experimentally measured yields; the same applies to energy and angular distributions. Sputtering is widely used for surface layer analysis, for removing surface atoms for thin film deposition and for surface machining.

1 Overview

If the surface of a solid is bombarded with energetic particles, it becomes eroded, surface atoms are removed, and the morphology of the surface gets modified. This phenomenon is usually named "Sputtering" in English and "Zerstäubung" in German. Sputtering was first discovered more than 150 years ago [1–7] as the erosion of the cathode in electric gas discharge tubes, and was named "cathode sputtering". Besides sputtering, several other effects are observed at particle bombardment of surfaces. A fraction of the incident ions is backscattered in collisions with target atoms [8–11], the others are implanted and come to rest in the solid in transferring their energy to electrons and lattice atoms [12]. After being slowed down they may diffuse further and finally be trapped in the solid or be desorbed at the surface [13,14]. The ion bombardment further causes the emission of electrons [15–20] and of photons [21–25] and, finally radiation damage in the surface layers of the solid, i.e. a change of the surface structure and topography [26–29].

It took about 100 years until the physical processes involved in sputtering were clearly identified [30,31] and about another 20 years until a quantitative description of the sputtering process for keV ions had been developed [32–39]. For energies in the 100 eV to the keV range sputtering is caused by atomic collisions between the incoming particles and the atoms in the surface layers of a solid, i.e. the incident particles initiate a collision cascade between the lattice atoms in the solid. For MeV ions also the energy deposited in electrons plays a major role for surface erosion. These processes are in principle the same as those causing radiation damage in the bulk of a solid [40–42]. The collisions take place far from thermal equilibrium, which means that sputtering with keV ions is generally different from evaporation. However,

evaporation contributes to sputtering for bombardment with MeV ions (see Chapter by Assmann, Toulemonde, Trautmann). A target atom is sputtered, if it has received an energy component normal to the surface larger than the surface binding energy. This energy is generally approximated by the heat of sublimation, which is smaller than the displacement energy necessary for creating a stable displacement (radiation damage in the bulk of a solid) [32–61]. These processes are named *physical sputtering*. They take place for all particle solid combinations if the incident particles have an energy above a threshold energy in the 10 to few 100 eV range. Sputtering may be enhanced or reduced, if the incident ions form a chemical binding with the atoms of the solid material. This process is named *chemical sputtering* [30]. If gaseous molecules are formed, sputtering is enhanced (see Chapter by Jacob, Roth), while at the formation of a stronger binding at the surface of the material such as an oxide or carbide, sputtering is reduced. The very large chemical sputtering yields of about 0.2 of carbon at bombardment with hydrogen ions were shown to be reduced by doping the carbon with metal powder [62,63] or fine grain carbide powder [64].

Depending on the energy of the incident particles and the collision cross-section between the incident particles and the atoms of the solid, for physical sputtering several collision regimes have been identified [46,61]:

The *single knockon regime* for light ions and low energy heavier ions. Here the recoil atoms from the incident particle-target atom collisions receive a sufficient high energy that they may be sputtered, but not enough energy to generate a recoil cascade.

The *linear cascade regime*: Here recoil atoms from the incident particle-target atom collisions receive a sufficiently high energy to generate recoil cascades. But the density of recoil atoms is sufficiently low so that knock-on collisions with atoms of the solid at rest dominate and collisions between moving atoms are infrequent.

The *spike regime*: This applies for incidence of heavy ions having a large collision cross-section and for the incidence of molecules or atom clusters. Here the density of recoil atoms is so high that the majority of atoms in a certain volume (the spike volume) is set in motion.

For bombarding energies in the MeV range additional processes such as *electronic sputtering* contribute to surface erosion. A major part of the energy of incident heavy ions is transferred to electrons along the ion track. A coupling between the electrons and the phonons causes large local heating in a cylindrical volume. Surface atoms may be removed by evaporation in a jet from the heated volume (see Chapter by Assmann, Toulemonde, Trautmann). For insulators this erosion process is much larger than for metals. Electronic sputtering has also been reported for the incidence of slow, highly charged ions [65]. In this case the large potential energy of the bombarding ions is dissipated into the electronic subsystem causing large sputtering yields for semiconductors and insulating materials [66,67].

For the different collision regimes the atoms sputtered have characteristic energy and angular distributions.

For crystalline targets, sputtering is largely influenced by the lattice structure. For ion incidence near close-packed crystal directions, the probability for collisions to create primary knockon atoms is reduced. Sputtering is lower than for other directions of ion incidence due to a higher transparency of the lattice and channeling [47,50–54,68,69]. The maximum of the energy dependence of the sputtering yield is shifted towards lower energies [53,56–60]. Furthermore, sputtered particles are preferentially emitted in close-packed crystal directions [48,70]. This has been attributed to focusing collision sequences [71–77]. However, the details of the last collision between the sub-surface layer and surface atoms can also play a major role [78].

In insulators, another energy transfer process is possible, which is also effective for low energy electrons and photons. The incident ions can produce a localized excitation which, after decay, creates a repulsive potential strong enough to replace lattice atoms [79]. The excitation in the bulk may diffuse to the surface and cause intense erosion [80].

2 The Sputtering Yield

The amount of erosion is measured by the sputtering yield, Y , which is defined as the mean number of atoms removed from the surface of a solid per incident particle:

$$Y = \frac{\text{average number of atoms removed}}{\text{incident particle}} \quad (1)$$

The definition of the sputtering yield is meaningful only, if the average number of atoms removed is proportional to the number of incident particles, which has indeed been found in most cases. Increased yields per atom are observed for heavy incident particles, such as molecular ions or atom clusters, at energies ≥ 10 keV, where spike effects [46,81–90] can cause a nonlinear dependence of the yield on the number of atoms per incident molecule or cluster [91]. The number of atoms sputtered for one incident particle generally largely vary, as shown in computer calculations [92–94].

The incident particles may be energetic ions or neutral atoms, molecules, larger atom clusters, such as fullerenes, neutrons [95], electrons or energetic photons [79]. For bombardment with monoatomic molecular ions, each incident atom is generally counted separately. In sputtering experiments with other molecules it may be appropriate to define the yield per incident molecule. In counting the atoms removed, only those from the solid are included, while incident particles which become implanted and later reemitted are not taken into account. This is different for selfsputtering, i.e., for bombardment with the same ions as the solid where this distinction cannot be made in experiments, but in computer programs. A selfsputtering yield of

unity means that on average, one atom is removed or reemitted per incident ion.

3 Distributions of Sputtered Particles

The particles are emitted with a broad distribution in energy, E_1 , in all polar exit angles, θ_1 , measured relative to the surface normal [96]. They are emitted predominantly as neutral atoms in the ground state with energies of a few eV, some are excited and generally less than 5% are ions at different charge states, q [97]. Y_q may be the total yield for atoms in state q with $Y = \sum Y_q$. The distributions of the sputtered particles are described by the differential sputtering yields

$$\frac{\partial Y}{\partial E_1} \quad , \quad \frac{\partial^2 Y}{\partial^2 \Omega_1} \quad (2)$$

In specific experiments, for example Secondary Ion Mass Spectroscopy (SIMS) [98], only sputtered ions within a given energy range and charge state ' q ' emitted into a given angular range are measured, i.e.,

$$\frac{\partial^3 Y_q}{\partial E_1 \partial^2 \Omega_1} \Delta E_1 \Delta \Omega_1 \quad (3)$$

is determined. The SIMS signal is generally proportional to the sputtering yield of the target species. However, the charge state q depends on the target composition and the bombardment conditions [97]. Therefore, calibration measurements are necessary.

The angular distribution of particles sputtered from single crystals shows maxima in close-packed crystal directions. For polycrystalline materials, the emission distribution is a superposition of the emission distributions of the differently oriented crystallites in the bombarded area. For normal incidence the angular distributions for amorphous and polycrystalline materials may be described in a first approximation by a cosine distribution. For heavy ions and low bombarding energies close to the threshold, more atoms are emitted at large angles, while for light ions and higher energies, more atoms leave the surface in the normal direction [96]. For oblique ion incidence the maximum of the emission distribution is shifted away from the incoming ion beam. In sputtering of compounds the different constituents may be sputtered with slightly different angular distributions [99,100].

4 Surface Topography

Surface atom removal by sputtering, ion implantation, trapping of the incident ions, and radiation damage in the surface layers generally cause a modification of the surface. This depends on the target material, its crystallinity

and temperature as well as on the ion species and their energy and angle of incidence [26–28,101,102]. In sputtering the surface is generally not uniformly eroded, differently oriented crystallites are eroded at different rates and on the surfaces of the crystallites steps and pyramids may develop (Fig.3) [103–105]. The surface roughness influences the sputtering yield. For bombardment with non-diffusing gaseous ions, blistering and flaking may occur at a fluence of 10^{17} to 10^{18} ions/cm², but will disappear at fluences of about ten times greater resulting in a very rough surface [27].

For bombardment with noble gas and reacting ions such as N_2^+ and O_2^+ in the energy range of 100 eV and a few keV besides nanostructuring also smoothing of the surface at an atomic scale is found [106]. Rough silver surfaces have been found to become polished by noble gas ion bombardment at normal incidence [107]. The surfaces of glasses and semiconductors may become polished at an atomic scale [106], as demonstrated in Fig. 1. Indium-containing III-V compound semiconductors, such as InAs, InP or InSb with initial rough surfaces get very smooth with a roughness of 0.2, 0.8 and 1.4 nm [108].

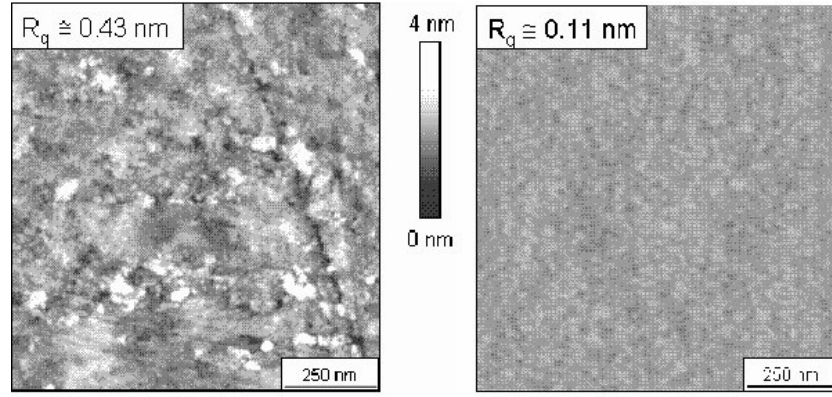


Fig. 1. Surface topography of fused silica; left: polished for optical purposes, right: after ion bombardment with 600 eV Ar at normal incidence, 300 nm material removed. The roughness has been reduced from 0.43 nm to 0.11 nm rms [109]

For low energy noble gas ions at normal or oblique incidence a regular ripple structure [110,111,113] and ordered nanoscale dots are observed. This has been explained by a competition of roughening due to sputtering and smoothing by surface diffusion [112]. An example is shown in Fig. 2.

If the target is rotated during the ion bombardment around an axis normal to the surface, also a periodic pattern of nanodots is formed [106,114–116].

For bombardment with noble gas ions with energies above a few keV larger grain boundaries, etch pits, steps and pyramids are found [26,103,117–119], such as shown in Fig. 3 [104]. Pyramids are also found, if a small amount of

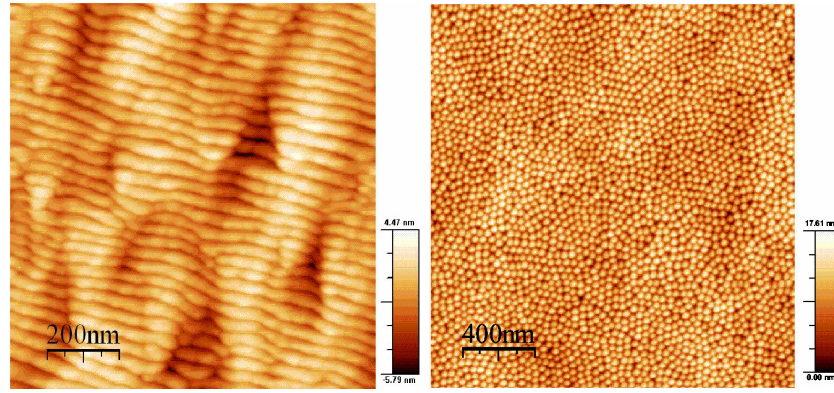


Fig. 2. Surface topography; left: ripple structure of Si bombarded with 500 eV Ar at an angle of incidence of 67° with respect to the surface normal after a dose of $1 \cdot 10^{18} \text{ cm}^{-2}$, right: dot structure after bombardment of GaSb with 500 eV Ar at normal incidence after a dose of $1 \cdot 10^{18} \text{ cm}^{-2}$ [113]

material with a low sputtering yield is evaporated onto the surface of a solid to be sputtered due to a shadowing effect [28,120,121].

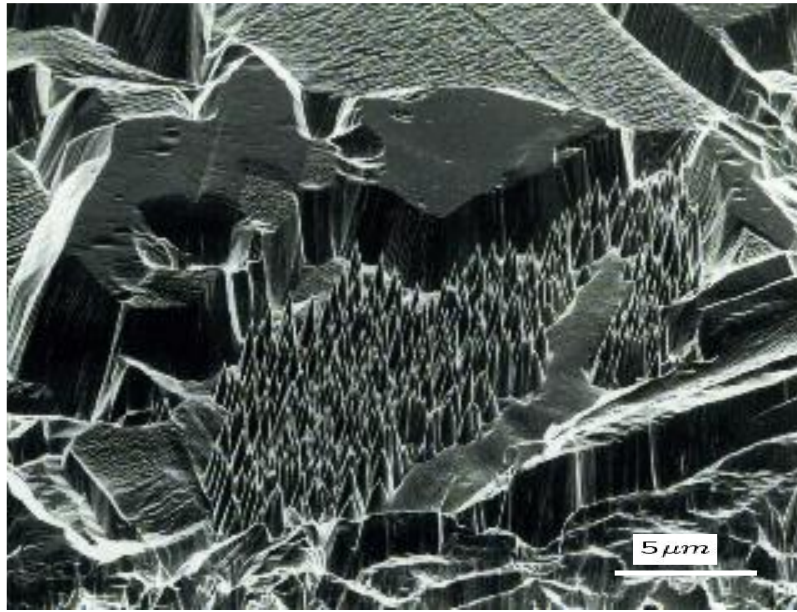


Fig. 3. Surface topography of polycrystalline Cu after 10 keV Kr bombardment at normal incidence and a fluence of 10^{18} cm^{-2} [104]

5 Sputtering Calculations

5.1 Analytic Theory

A major step forward in understanding the sputtering process for amorphous solids in the linear cascade regime was the application of integral transport equations [32,33,35–37,122–125] and their approximate solutions for a special potential ($V(r) \propto r^{-1/m}$ with $0 \leq m < 0.5$) for the atom-atom collisions and neglecting the energy loss to electrons by *Sigmund* [32,33,46]. First-order asymptotic solutions achieved for some parameter ranges resulted in an approximate algebraic formula for the dependence of the sputtering yield and the distribution of the sputtered particles on the bombardment conditions for an amorphous solid target of one element [32,33,46].

$$Y(E_0, \theta_0) = \Lambda F_D(E_0, \theta_0, 0) \quad (4)$$

Here E_0 is the incident energy and θ_0 the angle of incidence. $F_D(E_0, \theta_0, x = 0)$ is the nuclear energy deposition function in an infinite medium at depth $x = 0$, which is proportional to the nuclear stopping power $S_n(E_0)$ of the incident ion in the solid. Λ is a materials factor. For a planar surface barrier Λ is approximately given by:

$$\Lambda = \frac{\Gamma_m}{8(1-2m)NC_m U_s^{1-2m}} \quad (5)$$

with Γ_m being a factor of about 0.4 and C_m a factor in the nuclear cross-section [32,33,46]. Λ was further approximated by $\Lambda = 0.04/NU_s$ [126]. U_s is the surface binding energy, which is generally taken to be equal to the heat of sublimation. These equations show the dependencies of the sputtering yield on different parameters [32,33]. To include the behaviour close to the threshold energy Bohdansky has derived a new formula for the sputtering yield at normal incidence onto mono-atomic solids [127]. However, in this formula a too strong interaction potential (Thomas-Fermi) was used, and the formula gave too large threshold energies according to later calculated values [128], and it did not allow to fit the calculated values in a reasonable way [129].

For these approximations the differential yield for atoms sputtered in an energy interval ΔE_1 and an angular interval $\Delta \Omega_1$, see (3), is given approximately by:

$$\frac{\partial^3 Y \Delta E_1 \Delta \Omega_1}{\partial E_1 \partial^2 \Omega_1} = Y(E_0, 0) \frac{2}{\pi} (1 - 3m + 2m^2) \frac{U_s^{1-2m} E_1}{(E_1 + U_s)^{3-2m}} |\cos \theta_1| \Delta E_1 \Delta \Omega_1 \quad (6)$$

For $m = 0$ this reduces to

$$\frac{\partial^3 Y \Delta E_1 \Delta \Omega_1}{\partial E_1 \partial^2 \Omega_1} = Y(E_0, 0) \frac{2}{\pi} \frac{U_s E_1}{(E_1 + U_s)^3} |\cos \theta_1| \Delta E_1 \Delta \Omega_1 \quad (7)$$

The energy distribution of the sputtered particles has a maximum at an energy $E_1 = U_s/(2 - 2m)$ or, for $m = 0$ at half the surface binding energy. At high emerging energies ($E_1 \gg U_s$), the number of sputtered particles decreases proportional to E_1^{-2} . Deviations are observed for incident energies close to the threshold energy for sputtering, for oblique angles of incidence and especially for the situation of spikes.

5.2 Computer Calculations

Another approach to calculate absolute values for sputtering yields and the distributions of sputtered particles is to follow the collisions and the trajectories of incident ions and recoil atoms in amorphous or single crystal solids with computer programs [93]. Two different approaches have been used:

1. The *binary collision approximation* (BCA) which is realized in several computer programs, such as MARLOWE [47,130,131], TRIM (= transport of ions in matter) [132], TRIM.SP [34], TRIM-CASCADE [133], TRIDYN [38], and ACAT [134] (see Chapter by Eckstein, Urbassek).

2. *Molecular Dynamics programs* (MD) [135–137]. Here, after particle impact, the trajectories of all atoms in a solid are calculated taking into account the forces from all atoms on each other in some surrounding from their potentials (see Chapter by Eckstein, Urbassek). The advantage of the molecular dynamics method compared to the BCA approach is the possibility to calculate the formation of molecule and cluster emission. The disadvantage is the longer computational times and therefore a problem with good statistics.

Sputtering yields have been calculated for amorphous solids with TRIM.SP and ACAT for several incident energies and angles of incidence. The dependence of the sputtering yield on the incident energy as well as on the angle of incidence found in the calculations has been fitted with a new algebraic formula [129]. The sputtering yields measured by several authors are finally introduced into the new analytical curves (see Chapter by Eckstein).

Sputtering yields for polycrystalline solids with randomly oriented crystallites can be approximated to a first order by the yields calculated for amorphous solids. Thus the formulas derived for amorphous materials mostly give values in reasonable agreement with the sputtering yields measured for polycrystalline targets. For higher energies the binary collision approximation can be used, while for lower energies a molecular dynamics model may be more appropriate (see Chapter by Urbassek).

In the computer models it is possible to include an ideal or damaged surface and also damage in the solid. However, for these calculations a computer with a large memory is needed (see Chapter by Eckstein, Urbassek). The computer calculations are, however, especially useful for comparing the details of the calculated distributions of cascades with the assumptions made to achieve analytical solutions.

6 Sputtering Measurements

For the sputtering yields a large amount of experimental data has been collected, which have been measured under continuously improved conditions, i.e., with beams of mass analyzed ions having a well defined energy, in high vacuum at well characterized materials with flat surfaces [126] (see Chapter by Eckstein). Most of the experimental investigations of sputtering have been performed for mono-atomic, polycrystalline, single crystal and amorphous targets, mainly metals, at bombardment with noble gas and hydrogen ions. The sputtering yields depend on the mass, the energy and angle of incidence of the bombarding ions and the mass of the target atoms, on the surface binding energies, on the crystallinity and the orientation of the crystallites of the solid, but for amorphous and polycrystalline targets they are nearly independent of the target temperature [138–140]. For single crystals the minima in the sputtering yield at incidence in close-packed crystal directions become less pronounced with increasing temperature [141,142]. For magnetic materials the sputtering yield is modified according to the temperature dependence of the magnetization [143,144]. Below a threshold energy of about 5 to 400 eV for normal incidence, no sputtering takes place. Above this threshold energy the yields increase with increasing incident energy and reach a broad maximum in the energy region of 0.2 to 150 keV. (see Chapter by Eckstein). The decrease of the sputtering yields at higher energies is related to the larger penetration depth of the ions into the solid and a reduced energy deposition in the surface layers. Sputtering measurements with ions which react chemically with the atoms of the solid are still the field of many new investigations (see Chapter by Jacob, Roth) [145–148]. For bombardment with heavier ions of keV energies the yields are typically in the range of 1 to 10 atoms per incident particle, while for light ions the sputtering yields are well below 1. These results have been summarized and published in several review articles and books [22,30,43,44,48,61,93,126,149–165].

For ion bombardment at an oblique angle of incidence onto amorphous or polycrystalline solids, the sputtering yields increase monotonically with increasing angle of incidence up to a maximum around 50 to 80 deg., depending on the ion energy and the mass ratio between incident particles and target atoms, and it decreases again for larger angles of incidence. The location of the maximum depends further on the surface topography.

The sputtered particles are emitted predominantly as neutral atoms in the ground state and generally less than 5 % are ions [97]. A certain fraction can be emitted as atom clusters [136].

Sputtering can be observed in nature as well as in laboratory experiments. It occurs if matter in two extreme states, such as a hot plasma and a solid, interact with each other, or if a directed beam of energetic particles hits a surface. Such situations are found, for example, at the surface of the moon and other celestial bodies with no atmosphere, due to the impact of plasma particles from space such as the solar and the stellar winds [166–169]. Sputter-

ing is also found at the surface of space crafts and satellites especially during the movement in the very upper part of the atmosphere [170]. On earth energetic particles are produced by radioactive decay [171], in ion accelerators and at the boundary of plasmas. In laboratory experiments sputtering occurs, for example, at the cathode in electric gas discharges, where it was first observed. It is caused by energetic ions produced in the cathode fall [1–4]. Sputtering is also observed in the ion source of accelerators as well as at all diaphragms and targets hit by ion beams [172,173]. The effort to understand the sputtering process was also stimulated by a broad application in surface analysis, surface etching, cleaning, thin film deposition and the wall problem in fusion research [161,162,174–178].

Sputtering of alloys and compounds is still a field of many detailed investigations [64,146–148,179–181]. Generally, one component is initially removed at a larger rate so that the surface layer is enriched in the other component. At low temperatures where diffusion is suppressed, a steady state condition is reached where the atoms are finally removed stoichiometrically. However, at high enough temperatures, a depletion up to large depths in the solid is possible. These effects can influence the sputter deposition of thin films and the results of depth profiling measurements when using sputtering.

In the case of bombardment with ions, which do not react chemically with the atoms of the solid, a surface layer corresponding to the ion range will be damaged and some ions become trapped [31,50]. For bombardment with reactive ions, i.e. chemically enhanced or reduced sputtering, a compound surface layer generally builds up which has a different composition and structure than the material started with [145]. Sputtering effects will then correspond to those of the compound formed.

If the surface is contaminated such as by an oxide or carbide, these top atoms are predominantly removed and sputtering of bulk atoms can be largely reduced [50,182,183]. Generally, the effects involved with bombardment by species other than inert gas ions at different target temperatures are a field of more detailed studies.

The investigations of the differential sputtering yields are still very incomplete. However, these measurements, especially for atomically clean single crystal surfaces, allow to obtain detailed information about the sputtering process and the influence of the surface topography on the collision cascades.

7 Applications of Sputtering

Sputtering has long been regarded just as an undesired effect which destroys cathodes and grids in gas discharge tubes and ion sources. In high temperature plasma experiments with respect to controlled thermonuclear fusion, sputtering contributes to thinning of the vessel walls and a contamination of the hot hydrogen plasma with atoms of the vessel walls, which represents a

major problem in fusion research [174–177,184–188]. Sputtering is one of the causes for the destruction of diaphragms and targets in accelerators and in high-voltage electron microscopes [28]. In ion implantation the simultaneous removal of surface atoms by sputtering limits the achievable concentration in the implantation range [12].

However today, sputtering is used for many applications and has become an indispensable process in modern technology and physics [189]. Both the removal of atoms from a surface and the flux of atoms leaving the surface are successfully applied.

Sputtering allows a controlled removal of surface layers on a nearly atomic scale. It is applied for obtaining clean surfaces in field electron microscopy (FEM), field ion microscopy (FIM), low energy electron diffraction (LEED) and AUGER analysis of surfaces [189,190]. An intense ion beam with a geometrical sharp edge is used for nearly destruction free cuts for analysis of thick films such as in ion beam slope cutting [28]. The controlled erosion with well-focussed ion beams is especially useful for optical components, lenses [191–193] and even large mirrors of glasses and some insulators such as SiC [106,111,194,109]. The topography of the mechanically polished surface is analyzed with optical interferometry. The surface is finally machined and polished with a computer-controlled ion beam resulting in a submicron spatial resolution [195,196]. With noble gas ion beams in the 100 eV range or gas clusters with noble gas atoms at energies in the 100 eV range, the surfaces are polished or nanostructured on an atomic scale [106,195,197,198].

Sputtering is further applied for surface analysis and depth profiling of thin films. The ions removed by sputtering as well as the removed atoms after ionisation can be analysed in a mass spectrometer such as in SIMS or SNMS, and this gives very sensitive information about the surface concentrations, and a depth profile [199–202]. With rastered primary ion beams or imaging of the sputtered ions a high spatial resolution can be achieved [203–205]. If the surface is firstly smoothed by noble gas ion sputtering and sample rotation a very good depth resolution can be obtained [206,207].

One of the largest applications of sputtering is, however, the deposition of thin films on a large variety of substrates [208], onto large areas of several m² [209–214] or onto extremely small areas such as in microelectronics. The composition and properties of the deposited films depend on the deposition process, such as short or long pused or continuous sputtering of the substrate and the residual gas pressure during deposition [213].

8 Summary, Conclusions

Because the investigation of sputtering phenomena dates back more than 100 years, the field has been covered by a large number of review articles in many languages. Those which have appeared within the last 30 years give a vital

demonstration of the large advances made in understanding the sputtering process and its applications.

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