THE GROWTH DYNAMICS OF THICK SPUTTERED COPPER-COATINGS UNDER THE INFLUENCE OF SURFACE DIFFUSION: A QUANTITATIVE ATOMIC FORCE MICROSCOPY STUDY

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ABSTRACT:

The evolution of surface features during the growth of a coating is of vital importance for the resulting overall film structure and morphology. In the low substrate temperature regime (T/T_M<0.3) the growth front of a single component material roughens due to fluctuations in the incoming particle beam (kinetic roughening) and relaxes via surface diffusion. The continuous deposition process thus leads to the evolution of surface features of large lateral extensions compared to the extension of the film-forming particles.

In this paper a detailed experimental study of the surface evolution of sputtered copper coatings with thicknesses ranging from 300 to 10^5 Å for a working gas pressure of 0.4 Pa is presented. The roughening as well as the relaxation of the film growth front is studied by means of atomic force microscopy (AFM). The increasing lateral extension of the surface features during growth (coarsening) can well be described by a deterministic continuum model of surface diffusion. The model is capable to predict the evolution of the surface profile's Fourier transform for increasing deposition times (i.e. increasing film thickness). A re-transformation from k-space to real space allows for a direct comparison of surface profiles obtained from AFM-scans with those resulting from the continuum model and gives good qualitative agreement of the profile shapes.

1. INTRODUCTION:

The rapid development of techniques like scanning tunneling microscopy (STM) or atomic force microscopy (AFM) allows for a detailed study of surface processes on the atomic as well as on the mesoscopic and macroscopic level. A special interest has recently arisen in surface growth and erosion phenomena[1] which can now be quantitatively monitored by AFM. Eklund et al.[1] studied the dynamic evolution of the erosion of HOPG (Highly Oriented Pyrolytic Graphite) under energetic Ar^+-ion bombardment. These investigations resulted in the development of an equation of motion of the eroded surface which involves several types of transport mechanisms.

The same type of considerations can be applied to the process of film growth which is equivalent to the erosion process in the sense that particles are randomly added to a growing aggregate instead of removed. It is well known that whenever an initially smooth surface is exposed to a beam of incoming particles the growing interface begins to build up random fluctuations. In the early stages of growth the evolution of the amplitudes and frequencies of the surface fluctuations in space and time can be described by the mechanisms of kinetic roughening[2,3].
Lateron additional processes like surface diffusion and shadowing\textsuperscript{[4]} begin to affect the evolution of the roughness. The intention of this paper is to quantify the influence of surface diffusion processes on film growth under far-from-equilibrium conditions i.e. in the low substrate-temperature regime ($T_S/T_M \leq 0.3$) and for high deposition rates ($10^3$ Å/min). The growth process was monitored by investigating the surfaces of various samples with thicknesses ranging from a few 100 Å up to several µm. Other possible transport processes as e.g. evaporation/recondensation or viscous flow will be assumed as negligible. This assumption will be proven by experimental as well as theoretical results which show surface diffusion as the main mechanism of feature coarsening.

In Sec. 2 the deposition equipment is described and the experimental parameters are specified. In the following Sec. 3 we present the results of an AFM-investigation of the surface morphology of sputter deposited copper-films with thicknesses ranging between 300 Å and $10^5$ Å. The properties of the film-surfaces are characterized in respect to their RMS-values and Fourier-spectra.

In Sec. 4 quantitative expressions which relate the surface growth morphology to the amount of surface diffusion at a given substrate temperature will be developed. To accomplish this task a description of surface diffusion based on the continuum models of material transport along a surface derived by Herring and Mullins [5,6] is used. The theoretical expressions resulting from this model provide the possibility to predict the Fourier profile of the growing surface for a given deposition time. In addition a re-transformation from Fourier-Space to real space will be presented which allows the direct comparison of experimentally assessed surface profiles to the ones obtained from the model.

Sec. 5 provides a short summary and a discussion of the experimental and the theoretical results. Theory and experiment are briefly discussed in respect to the basic mechanisms of surface roughness evolution and the effects of mass transport along the surface on characteristic feature extensions. In conclusion possibilities of using the presented methods for the determination of effective surface mobilities in the active growth zone of a film during deposition are put to discussion. Future directions of research activities will be addressed.

### 2. EXPERIMENTAL SETUP:

All experiments were carried out in a turbomolecular pumped sputter plant (Alcatel SCM 450) equipped with a planar magnetron cathode of 100 mm diameter. The technical data of the sputter equipment are given in Table I.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Recipient diameter</td>
<td>450 mm</td>
</tr>
<tr>
<td>Recipient height</td>
<td>300 mm</td>
</tr>
<tr>
<td>Pumping speed (TMP)</td>
<td>450 l/s (Ar)</td>
</tr>
<tr>
<td>Base pressure</td>
<td>$10^5$ Pa</td>
</tr>
<tr>
<td>Pressure measurement</td>
<td>Baratron gauge</td>
</tr>
<tr>
<td></td>
<td>Penning gauge</td>
</tr>
<tr>
<td>Target diameter</td>
<td>100 mm</td>
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</tbody>
</table>

Table I: Technical data of the sputter equipment
Ar was used as working gas. The working gas pressure was set to a constant value of 0.4 Pa and monitored with a Baratron gauge during the deposition process. The sputtered material was Cu. The films were deposited onto atomically smooth oxidized Si(100) wafers mounted in a distance of 80 mm above the center of the magnetron source. For this pressure and distance the impingement angles of the particles arriving at the substrate are mainly determined by the erosion profile of the magnetron source are be limited to a maximum value of about 30° relative to the substrate normal [7]. Therefore self shadowing effects influencing the film growth mode are negligible. The deposition rate was held constant at 103Å/min. Samples were prepared with film thicknesses of 300 Å, 1000 Å, 3000 Å, 6000 Å and 105 Å. The experimental parameters are summarized in table II.

| Substrate material:                    | Si(100)-Wafer, oxidized, atomically smooth |
| Substrate Temperature:                | 450 K                                      |
| Substrates:                           | grounded                                   |
| Working gas pressure:                 | 0.4 Pa                                     |
| Working gas:                          | Ar                                         |
| Working gas pressure measurement:     | Baratron gauge                             |
| Target diameter:                      | 100 mm                                     |
| Distance target/substrate:            | 80 mm                                      |
| Deposited Material:                   | Cu                                         |
| Deposition rate:                      | 1000 Å/min (=17 Å/s)                       |
| Thickness of deposited samples:       | 300 Å, 1000 Å, 3000 Å, 6000 Å, 105 Å       |
| Deposition times:                     | 18 s, 60 s, 180 s, 360 s, 6000 s           |

Table II: Experimental parameters of the deposited Cu-samples

After deposition the samples were allowed to cool down to room temperature in the vacuum chamber. The samples were then removed from the vacuum chamber at room temperature to prevent oxidation due to elevated temperatures upon contact with the atmosphere and investigated by an AFM (Digital Instruments Nanoscope III) with a 500 Å radius Si(NO3)4 tip under atmospheric conditions. Several samples were once again investigated after prolonged storage (several days). The surface morphology was not influenced by the exposure to the atmosphere. Therefore we believe that oxidation does not alter the surface properties significantly. From the AFM-data the RMS-roughnesses and the Fourier-spectra of the surfaces were determined. A definition and a detailed description of the determination of surface Fourier spectra will be given in the next section.

3. EXPERIMENTAL RESULTS:

AFM-topographs of 5x5 µm were obtained from the deposited copper coatings were investigated in respect to roughness evolution and the evolution of the surface Fourier-spectra. The RMS-roughness (surface width) is defined by the expression

\[
\xi^2 = \frac{1}{N} \sum_{i=1}^{N} (h_i - \bar{h})^2
\]  

(1)
for a system containing N sites with single valued heights \( h_i \). \( \bar{h} \) in eq. (1) denotes the average height of the system, the \( h_i \) are measured relative to \( \bar{h} \). For the theory presented in the next section will deal only with \textit{single one dimensional surface scans} the RMS-values were determined in a slightly modified way: the \( \xi \)-values were determined for a set of about 50 linear surface scans of 5 µm length arbitrarily located in the AFM topographs. The \( \xi \)-values obtained this way were then averaged to obtain an estimate of the variance of the RMS-value of \textit{one single linear scan} in a topograph. Fig. 1 shows a log/log-plot of the dependence of the RMS-value on the deposition time of the film. The error bars in fig. 1 result from the averaging procedure.

![Log/log-plot of the roughness evolution of Cu-films with increasing deposition time. The error bars result from averaging the RMS-values of several arbitrary 5 µm long surface scans of the AFM-topographs.](image)

Fig. 1.: Log/log-plot of the roughness evolution of Cu-films with increasing deposition time. The error bars result from averaging the RMS-values of several arbitrary 5 µm long surface scans of the AFM-topographs.

The log/log-plot in fig. 1 shows a linear characteristic with a growth exponent for \( \xi \) corresponding to

\[
\xi \propto t^{\beta}
\]

with \( \beta = 1/3 \) as can be seen from the function fitted to the data points. This corresponds to the growth class of the KPZ-equation for a one-dimensional interface\(^2\). As it is visible from Fig. 2 the roughening process is present during the whole considered set of deposition times. No smoothening is observed. Nonetheless an analysis of the roughening behavior alone does not give a complete description of the surface evolution because it tells nothing about the nature of lateral correlations on the surface which should be significantly influenced by surface diffusion.

To obtain informations about lateral surface correlations the AFM-height data were Fourier transformed along 5 µm long scans through the central point of the AFM topograph according to

\[
h(x) = \sum_n h_n(k_n) \sin(k_n x + \varphi_n).
\]

To obtain a circular average of the Fourier spectra the scan lines were rotated and the coefficients \( h_n(k_n) \) were averaged. The phase factors \( \varphi_n \) can be assumed as random. With this procedure it is possible to reduce the 2d-surface Fourier transform to one dimension given the assumption that the surface is isotropic in any direction. Since the surface roughness increases
with increasing deposition time, the absolute intensity of each single Fourier mode will increase. To allow a direct comparison of the relative intensities of the single modes in spectra for different deposition times, the spectra thus were normalized to their maxima close to \( k = 0 \). The spectra obtained after the described steps are shown in fig. 2.

![Fig.2.](image)

From fig. 2 it is clearly visible that the dominant modes concentrate near a value of \( k = 0 \) with increasing deposition time, i.e. that only long wave fluctuations will survive. The quantification of this behavior by a simple theory involving mass transport along the surface by surface diffusion is the aim of the next section.

4. THEORY:

This section shall elucidate the mechanisms related to surface diffusion, which govern the evolution of the lateral extension of surface features. We will not focus on the roughening behavior which is related to the shot noise of the incoming particle beam. Where an explicit knowledge of the functional dependence of the RMS-roughness \( \xi \) on deposition time is needed we will refer to the experimental data presented in the previous section.

4a. The influence of surface diffusion on the surface Fourier transform:

The averaging procedure involved in the determination of the surface Fourier transform in sec. 3 reduced the 2d surface Fourier transform to a one-dimensional Fourier-spectrum. Therefore the theoretical considerations about surface diffusion will be limited to the case of a one-dimensional Fourier-spectrum and therefore to the case of a one-dimensional interface.

At every finite substrate temperature \( T_s \) surface diffusion tends to smooth features with extensions below the diffusion length of an adatom in the film growth front. To quantify the influence of surface diffusion on the surface evolution of a coating it is possible to apply a continuum model which was first proposed by Herring\(^5\) and Mullins\(^6\). The most important consequence of these calculations in respect to thin film growth is that features with small amplitudes are unstable against surface diffusion. Due to the material transport triggered by
surface diffusion the normal growth velocity for a sinusoidal profile with the amplitude \( A_0 \) and the wavenumber \( k \) at the maxima, \( v_D^{\text{MAX}} \), and at the minima, \( v_D^{\text{MIN}} \), is given by

\[
\frac{v_D^{\text{MAX}}}{v_D^{\text{MIN}}} = -\frac{D_S \Omega^2 \nu}{k_B T_S} A_0 k^4 = -v_D^{\text{MIN}}. \tag{4}
\]

In eq. (4) \( D_S \) denotes the surface diffusion coefficient, \( (D_S = D_{S0} \exp[-E_D / k_B T_S]) \), \( \gamma \) is the surface tension, \( \Omega \) the atomic volume of the diffusing species, \( \nu \) gives the number of diffusing atoms per unit area, and \( A_0 \) is the Amplitude of the undulation. If these material constants are known one can calculate the normal growth velocities given in eq (2) for an arbitrarily chosen mode of a Fourier spectrum.

Furtheron the dependence of growth velocities on the wavelength of a surface fluctuation provides the possibility to study the influence of surface diffusion on a profile generated by a random deposition process. If it is possible to include a time dependence into the Fourier-expansion of the surface profile according to

\[
h(x, t) = \left[ \sum n h_n(k_n) \sin(k_n x + \phi_n) \right] \zeta(t) = \left[ \sum n h_{n0}(x) \right] \zeta(t) \tag{5}
\]

then the evolution of the single Fourier components in time is described by an exponential characteristic of the form\[6\]

\[
h_n(x, t) = h_{n0}(x) e^{-AF_n t}, \tag{6}
\]

where \( F_n \) is the frequency of the \( n^{\text{th}} \) Fourier component given in full oscillations per \( \mu \text{m} \) and the material dependent constant \( A \) is given by

\[
A = \frac{\gamma \Omega^2 \nu}{k_B T_S} D_{S0} e^{\frac{E_D}{k_B T_S}} (2\pi)^4 10^{16}. \tag{7}
\]

Inserting the material constants for \( \text{Cu} \) given in table III, \( A \) has a numerical value of \( 1.46 \times 10^{-6} \mu \text{m}^4 \text{s}^{-1} \).

| Pre-exponential of \( D_S, D_{S0} \): | 1.4x10^{-2} cm²s⁻¹ |[8] |
| Activation Energy \( E_D \): | 1.21x10⁻¹⁹ J |[8], |
| \( D_S \) at 450 K: | 4.2x10⁻¹¹ cm²s⁻¹ |
| Surface tension \( \gamma \): | 1.28x10⁻⁴ Jcm⁻² |[9] |
| Atomic volume \( W \): | 8.6x10⁻²⁴ cm³ |
| Number of diffusing atoms in the surface \( \nu \): | 1.5x10¹⁵ |

Table III: Material parameters of \( \text{Cu} \) entering eqs. (4, 7, 8)

The Fourier spectrum, which is defined as the plot of the coefficients \( h_n \) in dependence on \( F_n \) (i.e. the relative Fourier intensity, if the \( h_n \) are normalized to their maxima) will then also
follow an exponential decay law because the coefficients \( h_n(F_n) \) can be redefined as \( h_n(F_n, t) \) according to

\[
h_n(F_n, t) = h_n e^{-AE^2 t}.
\] 

(8)

Assuming the initially deposited surface as random, the initial spectrum can be taken as a constant for frequencies form \( 0 \rightarrow 1/a \), where \( a \) is the linear extension of the film-forming particles\(^1\). Immediately after \( t=0 \) the high frequency components will decay rapidly according to eqs. (4,8). Fig. 3 shows calculated Fourier-spectra at different deposition times for copper deposited at 450K.

![Fourier spectra calculated from eq. (8). The experimentally observed concentration of the relative intensities towards low Frequencies with increasing deposition times is well reproduced.](image)

The calculated spectra can be compared with Fourier spectra obtained from the AFM measurements as it is done in Fig. 4. As it can be seen from Fig. 4, there is reasonable agreement between the calculated and the measured spectra. The slower decay towards higher frequencies in the measured spectra can be attributed to the permanent noise in the incoming particle beam which continuously generates new small wavelength features during the whole deposition process.

![Direct comparison of measured and calculated spectra. The presence of high frequency modes in the measured spectra can be attributed to the noise in the deposition beam which continuously generates short wavelength features and which was completely neglected in the calculations.](image)
The half-width of the spectra can be used as a measure to estimate the variation of the mean lateral extension of surface features with deposition time. The dependence of the half-width $F_H$ (frequency where the spectra derived from eq. (8) have a value of 0.5) on the deposition time is given by the expression

$$F_H(t) = \sqrt{\frac{-\ln(1/2)}{A}}(t)^{-1/4}$$

(9)

and is displayed in Fig. 5 where the black squares represent the half widths of the spectra determined from the AFM-data.

![Fig. 5: Comparison of measured and calculated half widths of the spectra. The half width is systematically overestimated in the calculation. This could be a result of the uncertainty in the diffusion data for Cu.](image)

As can be seen from Fig. 5 the rapid decay of small surface features for short deposition times is well reproduced by eq. (7), although the experimental half widths are systematically lower than the calculated ones. Fig. 5 also shows that the lateral extension of surface features will only vary slowly for deposition times greater than $10^4$ s. This fact indicates a situation where the lateral feature extension reaches a sort of a saturation value.

Considered the simplicity of the model and the fact that eq. (8) contains no free parameters calculation and experiment coincide well. Especially the variation of the half-width of the spectra is quantitatively reproduced by the model over a large range of deposition times.

4b: Retransformation into real space:

The calculations performed in Sec. 4a allowed the construction of deposition-time dependent one-dimensional surface Fourier-spectra. It is now possible to pose the question, if one can use the information from the calculated spectra and re-transform them into real space. In fact, such a re-transformation can be performed under two assumptions which result from the presence of the deposition noise:

- The influence of the deposition noise results in a randomization of the phases which are the second parameter determining the actual shape of the surface profile besides the relative intensities of the Fourier modes.
- The roughening behavior, which is not described by spectra containing only relative Fourier intensities has to be obtained from different measurements.
Also it seems to be necessary to make the remark that such a retransformation only yields a one-dimensional surface profile since the measured Fourier profiles contain only a radial average of the surface-Fourier transform and the calculated surface profiles are obtained from a one dimensional theory.

Bearing these facts in mind the theoretical Fourier-Spectra can be transformed to real space according to

\[ h(x,t) = \sum_n h_n(F_n,t) \sin(2\pi F_n + \varphi_n) \]  

(10)

\[ \varphi_n = \text{random phase factors, } \varphi_n \in [0,2\pi]. \]

After performing the re-transformation according to eq. (10) and matching the RMS-values to the experimental data, the profiles obtained this way can be compared with measured profiles of arbitrary AFM-surface scans. The results of the re-transformation for different deposition times are shown in Figs. 6a-d, AFM-scans of films prepared with the same deposition times are displayed in Figs. 7a-d. There is good qualitative agreement in the real space morphology. The good agreement between experimental and theoretical results makes it possible to approach different aspects of the growth of thick films. These aspects shall briefly be addressed in the following concluding section.

Fig. 6.: Surface profiles obtained from the re-transformation in eq. (10). The Fourier spectra entering eq. (10) were derived from eq. (8) for the deposition times displayed in the profile graphs. Finally the RMS-values of the theoretical profiles were matched to the experimental values assessed from Fig. 1
Fig. 7.: Measured surface profiles determined from the AFM topographs for samples of different deposition times.

5. DISCUSSION AND CONCLUSION:

The influence of surface diffusion on a growing surface profile of a Cu film was investigated by AFM and quantitatively described using a simple continuum model based on the assumptions of Herring\cite{5} and Mullins\cite{6}. The experimental and theoretical results can be summarized in three important points:

- Up to great film thicknesses the RMS-roughness increases according to a power law as it is familiar from kinetic roughening theories. There is no smoothing effect by surface diffusion in the global behavior of the RMS-value during film growth.

- Concerning the lateral extension of surface features, the film’s growth front behaves like a free surface under the influence of surface diffusion. Features of small lateral extension decay rapidly. This process can be monitored by comparing the relative intensities of the surface modes present in the Fourier-spectra of films of various thicknesses. The decay of small wavelength features does not seem to be affected critically by the presence of the incoming particle flux as far as the relative Fourier-intensities of the various surface modes are concerned. Therefore the relative intensities of surface modes in all stages of the deposition process can be described by a model of the evolution of a free surface which is subjected to a relaxation via surface diffusion.

- The Fourier-Spectra obtained from the theory can be re-transformed into real space. The one dimensional surface profiles are qualitatively similar to those obtained from arbitrary surface scans of the AFM topographs.

The asymmetry in vertical and lateral surface evolution indicated in the first two points can be ascribed to the asymmetric dependence of the diffusion fluxes on the vertical and the lateral extension of an arbitrary surface feature (for this fact see eq. (4)). While diffusion fluxes vary with the fourth power of the wavelength of a surface feature, the vertical extension of the feature affects the amount of the diffusion flux only linearly. Therefore *roughening*,
which is mainly the variation of the vertical extension of surface features, is only weakly affected by surface diffusion. On the other hand coarsening (i.e. the evolution of lateral feature extensions during the growth process) can quantitatively be described by a pure surface diffusion approach which completely neglects the presence of the incoming particle flux.

The predictability of the surface profile mentioned in point three allows the a semiquantitative determination of the extension of characteristic surface features for all phases of growth. This fact is important since the knowledge of characteristic feature sizes and shapes provides informations about the further film growth mode. A pronounced surface profile with characteristic features which is subjected to a broad angular distribution of impinging particles can e.g. be the nucleus of columnar film growth via the shadowing growth instability\[4,10-12]\. A detailed knowledge of the profile evolution with deposition time can therefore give information about the onset of columnar growth in dependence on deposition time. A study of this problem will be subject to further work.

Another aspect of the interpretation of the experimental data is the determination of effective surface diffusivities by fitting the curves from eq. (8) to experimental data as e.g. to the half width $F_H$ of experimentally determined spectra (which was systematically overestimated by eq. 9). This procedure would provide surface diffusion data of a certain material under actual growth conditions which should be of considerable interest for many technological applications. For this task it is of course necessary to extend the data set beyond the data provided in this work and to further justify the theoretical approach by performing more experiments with different deposition rates and substrate temperatures. Additional work in this direction as well as a variation of the deposition geometry from the planar to a post magnetron geometry is planned for the near future.

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REFERENCES: