

The influence of surface roughness on electronic transport in thin films

G. Reiss and H. Brückl

Institut für Angewandte Physik, Universität Regensburg, Universitätsstrasse 31, D-8400 Regensburg, Germany

Received 9 September 1991, accepted for publication 10 September 1991

In thin films, the structure of the surfaces considerably influences the transport of conduction electrons. For mesoscopic roughnesses in the range of a few nm, this is due to the varying film thickness, which gives rise to a spatially fluctuating conductance. Moreover, microscopic roughnesses can contribute to the scattering of the electrons and therefore additionally enhance the thin-film resistivity. For a quantitative understanding of the transport in these systems, a detailed investigation of the surface roughness combined with measurements of the electronic properties are necessary. Here, we discuss STM imaging of various metal films and the application of these results to the interpretation of electronic thin-film properties. Provided reasonable resolution of STM in the nm range, a good correspondence of STM results with the electrical behaviour of growing metal films can be established. Furthermore, a detailed two-dimensional analysis allows for a calculation of the potential on current-carrying thin films. On the other hand, this method supplies reliable values for the electronic transport parameters.

1. Introduction

The conductivity of thin films has been frequently discussed in the literature. Whereas semi-classical models used the Boltzmann equation with appropriate boundary conditions [1], more recent contributions [2-4] gave quantum-mechanical models especially for the thickness dependence of the conductivity. In these discussions, special regard must be naturally taken on the surface scattering of the conduction electrons which is the main reason for the increase of the resistivity of thin films. In ref. [4], it was suggested, that a possible variation of the electron density with thickness could additionally influence the conductivity.

The main purpose of these discussions is the evaluation of reliable transport parameters from the thickness dependent conductivity $\sigma(d)$ (d : film thickness). In order to obtain these values, the theoretical curve must be fitted to the experiment by a variation of usually at least three parameters, one of them representing the surface

profile. This, however, usually was performed in one dimension without knowledge of the real surface. Therefore, a detailed STM analysis of the thin-film surfaces can considerably improve these investigations. Since STM directly images the surfaces, the discussion of the conductivity can be extended to two dimensions. With this treatment the distribution of the electrostatic potential on current carrying thin films can be additionally estimated. The precondition for these evaluations, however, are reliable results of STM imaging.

2. Surface imaging

The imaging of the thin-film surfaces was performed with STM under ambient conditions [5]. The materials have been Pt, Au, Ni, Co and Cu. In order to evaluate possible influences of oxidation, Ni and Cu was partly covered with a 1-2 nm thick Au overlayer. In the case of Ni, STM revealed identical surface features without remark-

able signs of oxidation on the unprotected samples. On pure Cu, reliable imaging was possible only for a few hours after the removal of the films from the UHV chamber. The Cu films protected by Au, however, exhibited the same surface features and allowed stable tunneling for considerably longer time.

As mentioned in the introduction, the resolution of nm features is of special interest for an application of STM results to electronic transport. This point has been already stressed in refs. [6,7]: based on the STM image and a properly evaluated tip shape, the real surface can be reconstructed except those parts, the tip was not in tunneling contact with during scanning the surface. The numerical procedure proposed in ref. [8] has been recently reformulated in terms of Legendre transforms [9]. In the recalculated image, the parts which had not been reached by the tip remain as "black holes". Since the amount of black holes obviously is directly related to the obtained height resolution, we show in fig. 1a a model surface consisting of semi-elliptical islands. If the tip indicated in fig. 1a scans this surface, the resulting STM profile corresponds to the line shown in fig. 1b. Using the definitions of the height resolution indicated in fig. 1, the dependence of this quantity on the amount of surface without tunneling contact to the tip can be estimated. The result of this calculation is shown in fig. 2 for different ratios $a = R/H$.

As can be seen from this estimate (fig. 2), the obtained height resolution depends very critically on the amount of unresolved surface area. Only a few percent of black-hole surface can cause a considerable underestimate of the roughness. For the further discussion, we therefore used only

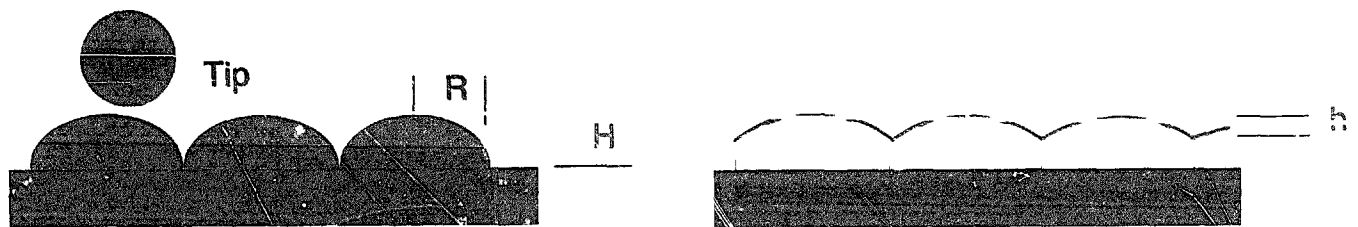


Fig 1 (a) Model surface for estimating the height resolution of STM (b) STM profile corresponding to the tip/surface combination of (a). The height resolution is defined as the ratio h/H

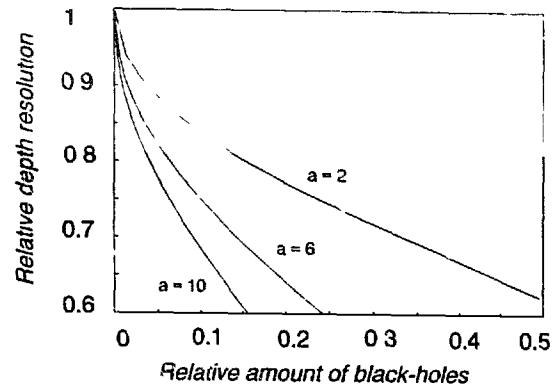


Fig. 2 The height resolution of STM images as a function of the surface area without tunneling contact to the tip

images with a black-hole area equal or less than 1% of the whole surface.

If, however, the precondition concerning the resolution is fulfilled, STM clearly can provide reliable surface profiles for the discussion of the electronic transport properties.

3. The thickness dependence of the resistivity

For the evaluation of the transport parameters, we use the model of Tešanović et al [2]. Here, the thickness dependence of the conductivity is given by:

$$\sigma_{loc}(d(x), l_x h^2, \sigma_c) = \frac{\sigma_x}{n_c} \sum_{n=1}^{n_c} \left[1 + \frac{l_x h^2 k_f^2}{6\pi d(x)} \left(\frac{n}{n_c} \right)^2 \right]^{-1}, \quad (1)$$

where the sum includes the occupied subbands (index n), $d(x)$ is the local film thickness, k_f is the Fermi wavevector, σ_x is the conductivity of

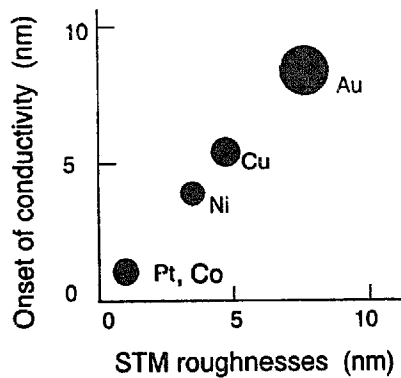


Fig. 3. Onset thickness of the ohmic conductivity of films growing on glass substrates at room temperature compared with the corresponding roughness found by STM. The radius of the dots corresponds to the spread of the experimental data

infinitely thick material and $l_x h^2$ is the product of the intrinsic electronic mean free path and the microscopic roughness of the surface. Roughnesses considerably larger than the Fermi wavelength can be included by averaging over a varying film thickness $d(x)$ similar to the discussions given in refs. [4,7,10].

The application of STM results can be performed in one dimension simply by using a corresponding one-dimensional roughness distribution. The preconditions and results of this treatment have been already discussed in ref. [7] for thin Ni films. In order to demonstrate the relevance of the method for different materials, fig. 3 shows the correlation of roughnesses estimated by two methods:

First, the roughness can be simply obtained via highly resolved STM images. Second, the mean roughness should correspond to the thickness of the growing film, where the onset of the ohmic conductivity can be found [7] (see eq. (1)).

From fig. 3, a surprisingly good agreement between these values of the thin film roughness can be established for different metals in a range from about 1 to 10 nm.

On the other hand, the one-dimensional model cannot reproduce the conductivity especially for very small thicknesses [7]. This is due to the lack of percolation, i.e., within this treatment the conductivity approaches zero at a thickness corresponding to the maximum corrugation found by

STM. In order to achieve a better description, we thus extended our discussion to two dimensions.

For this purpose, the film was modeled as a (128×128) networks of resistors, each of which represents a small portion of the integral film resistance. The corresponding local conductances can be obtained from eq. (1) in combination with the local thicknesses supplied by the STM image. In order to obtain the conductivity, the voltage has to be fixed at two edges of the STM image. The resulting voltages V_i at node i of the network can be obtained selfconsistently using the local conductances g_{ij} between node i and node j [11]:

$$V_i = \sum_{j=1}^4 \frac{V_j}{g_{ij}} / \sum_{j=1}^4 g_{ij}. \quad (2)$$

This self-consistent formalism quickly converges towards the distribution $\{V_i\}$ of the potential on the thin-film surface. Using this potential, the current can be evaluated using the local conductances defined before. From these values of current and voltage, the integral conductivity can be obtained in the usual way.

In fig. 4, we show the results of this two-dimensional estimate together with the simpler one-dimensional model and the experimental results for the resistivity of a Ni thin film

As can be seen from fig. 4, the one-dimensional fit approaches the experimental results at a thickness of about 7 nm. The maximum depth of

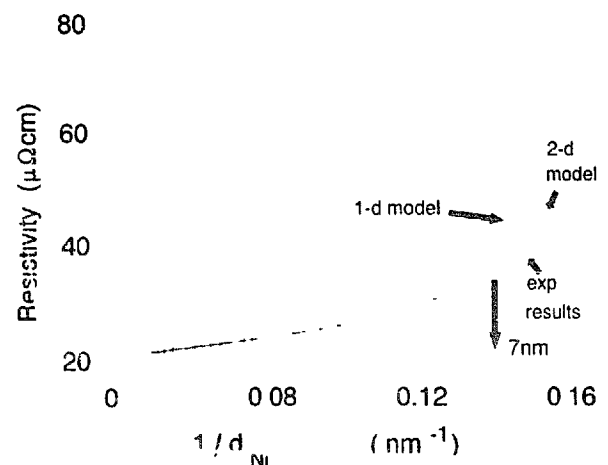


Fig. 4 Results of fitting the theoretical expressions for the thickness-dependent thin-film conductivity to the experimental curve

the bumps found by STM on this surface was about 6 nm [7]. On the other hand, the two-dimensional treatment can reproduce the experimental curve down to much smaller thicknesses. As already mentioned, this is due to the percolative behaviour for thicknesses smaller than the maximum roughnesses, which can be treated solely by a two-dimensional description.

The bulk resistivities ρ_x obtained from the fitting calculations are 19.5 and 21 $\mu\Omega$ cm for the one- and two-dimensional case, respectively. For the product of the mean free path with the microscopic surface roughness, we obtained 1.25 nm³ (1D) and 1.4 nm³ (2D). Since the two-dimensional treatment is more realistic, the mean free path therefore seems to be somewhat larger than the values obtained from the one-dimensional discussion. Nevertheless, already the simple one-dimensional model reveals rather correct values for this fundamental transport parameter.

The two-dimensional model, however, can supply more information: As discussed before, the distribution of the electrostatic potential on the current-carrying thin film must be calculated for fitting the theory to the experiment. On the other hand, this distribution itself is of considerable

interest. In fig. 5 we show the original STM topography of a 10 nm thick Ni film (fig. 5a) and the corresponding calculated distribution of the local electrostatic field defined as the magnitude of the gradient of the local potential. A good correspondence of the topographical features with the local field can be established: at locations of small film thickness, the local field can be enhanced by a factor two compared with the mean field.

Fig. 5 therefore shows, that a rather inhomogeneous distribution of the potential on rough thin films can be expected due to the spatially varying conductances. On the other hand, the drop of the potential itself is not as inhomogeneous as found by potentiometric STM measurements on polycrystalline thin films [12,13]. Thus, it seems to be necessary to include grain boundary scattering in the discussion of the experimentally evaluated potential [14].

4. Conclusions

In this contribution, we presented a two-dimensional treatment of the thickness-dependent

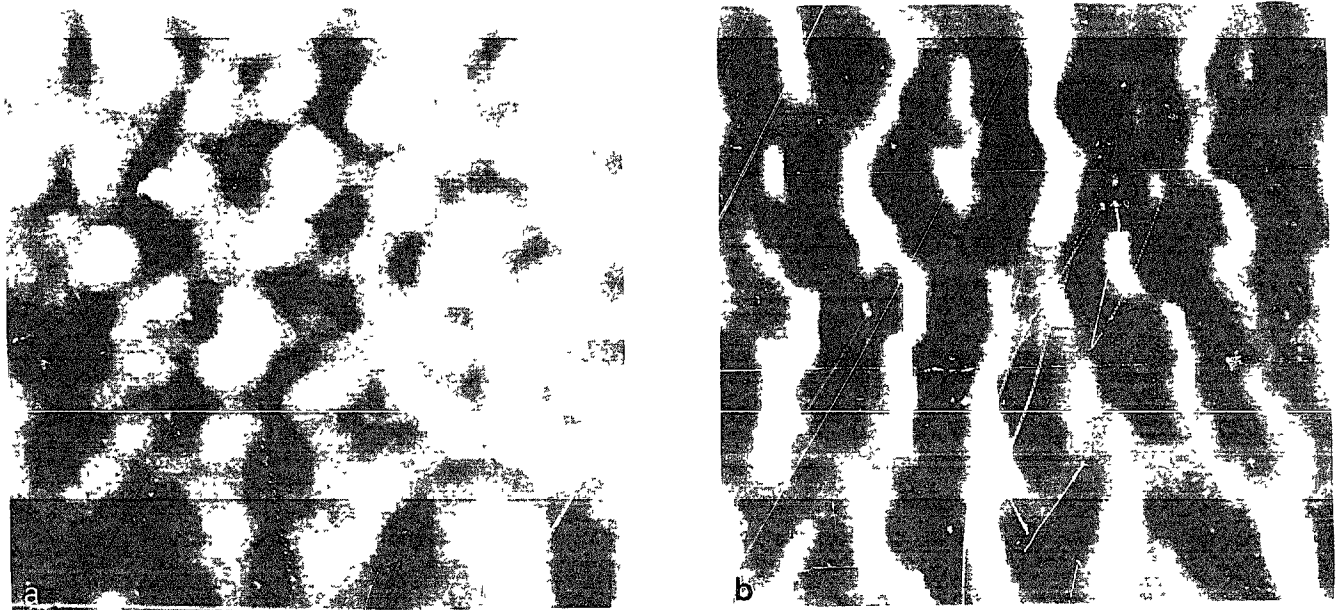


Fig. 5 (a) Grey-scale image of the topography of a 10 nm thick Ni film (180 nm \times 180 nm). The scale is 10 nm from dark to white. (b) The magnitude of the local field calculated for the surface topography shown in (a). White corresponds to a field twice as large as dark.

thin-film resistivity. The surface profiles necessary for this purpose can be supplied by highly resolved STM images. The STM roughnesses of the surfaces have been shown to correspond very well with the thickness of the onset of ohmic conductivity, i.e., just with the stage of coalescence of the growing film.

A two-dimensional fit of the experimental data of the thickness-dependent thin-film resistivity can be performed using a selfconsistent resistor network model and the topography supplied by STM. Although the realistic two-dimensional model supplies slightly different values, the results for the transport parameters agree rather well with former one-dimensional treatments [7,10]. In contrast with this, however, the extended discussion can include the stage of percolation, i.e., using this formalism, network structures can be treated. Moreover, this formalism naturally supplies the distribution of the potential on a current-carrying thin film. This turns out to be rather inhomogeneous as soon as the roughness becomes comparable to the film thickness. The potential obtained, however, is still smoother than observed by potentiometric STM measure-

ment. Therefore it seems to be necessary to include additionally grain boundary scattering in order to explain these results.

References

- [1] K Fuchs, *Proc Cambridge Phil. Soc.* 34 (1938) 100.
- [2] Z Tešanović, M V Jarić and S. Maekawa, *Phys. Rev Lett* 57 (1986) 2760
- [3] D. Calecki and G Fishman, *Surf Sci* 229 (1990) 110.
- [4] N Trivedi and N W Ashcroft, *Phys. Rev B* 38 (1988) 12298.
- [5] J Vancea, G. Reiss, F. Schneider, K Bauer and H Hoffmann, *Surf. Sci* 218 (1989) 108.
- [6] G. Reiss, H Brückl, J Vancea, R. Lecheler and E. Hastreiter, *J. Appl Phys.* 70 (1991) 523
- [7] G. Reiss, E. Hastreiter, H. Brückl and J Vancea, *Phys. Rev. B* 43 (1991) 5176
- [8] G. Reiss, F. Schneider, J. Vancea and H. Hoffmann, *Appl. Phys Lett* 57 (1990) 867
- [9] D. Keller, *Surf Sci.* 253 (1991) 353
- [10] U. Jacob, J Vancea and H. Hoffmann, *Phys. Rev B* 41 (1990) 11852
- [11] S. Kirkpatrick, *Rev. Mod Phys* 45 (1973) 574
- [12] J P Pelz and R H Koch, *Rev. Sci Instrum.* 60 (1989) 301
- [13] J P Pelz and R H. Koch, *Phys Rev B* 41 (1990) 1212
- [14] C S Chu and R S Sorbello, *Phys Rev B* 42 (1990) 4928.