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1989 J. Phys.: Condens. Matter 1 9867

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J. Phys.: Condens. Matter 1 (1989) 9867–9873. Printed in the UK

A new method for determining the electronic mean free path in polycrystalline metals

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Received 22 March 1989, in final form 11 July 1989

Abstract. In thin metallic films, both surface and volume scattering of the conduction electrons (CEs) contribute to the resistivity. The electronic mean free path (MFP) therefore can be evaluated by a separation of these two scattering mechanisms. In this paper we discuss a new and simple method for this. Owing to enhanced surface scattering, coating of thin films leads to an increase in the resistivity. From the relative change in the resistance during coating at different temperatures the MFP of the CEs can be evaluated. Moreover, surface coating experiments combined with measurements of the thickness-dependent conductivity additionally provide accurate values of the electrical transport parameters in polycrystalline metals.

1. Introduction

The evaluation of the electronic mean free path (MFP) in thin metal films is a longstanding problem of metal physics. For thin films the electrical resistivity is enhanced by diffuse surface scattering. Therefore the MFP can be determined by an analysis of the thickness-dependent resistivity.

The basic theory treating the thickness-dependent resistivity $\rho(d)$ (where d is the film thickness) was given in [1]:

$$\frac{\rho_\infty}{\rho(d)} = 1 - \frac{3(1-p)}{2k}\int_1^\infty \mathrm{d}t \left(\frac{1}{t^3} - \frac{1}{t^5}\right)\frac{1-\exp(-kt)}{1-p\exp(-kt)} \tag{1}$$

with ρ_∞ the resistivity of a very thick film with the same structure as the film under discussion, l_∞ the corresponding MFP of the conduction electrons (CEs), $k = d/l_\infty$ and p the specularity parameter. For $l_\infty \gg d$, equation (1) can be approximated by [2]

$$\rho(d)/\rho_\infty = 1 + (3l_\infty/8d)(1-p). \tag{1a}$$

The determination of the MFP by fitting equation (1) or (1a) to typical experimental values is possible only for films with thicknesses larger than approximately 10 nm. For thinner films the theory cannot describe the experiment [3]. Therefore (*a priori*) assumptions have been introduced. Commonly the assumption

$$\rho_\infty l_\infty = \rho_b l_b \tag{2}$$

(where $\rho_b l_b$ is the value for an ideal (bulk) single crystal, i.e. only phonon scattering is

taken into account) has been used [4]. Within a free-electron model, this product depends only on the density of CEs.

Besides surface scattering the interaction with grain boundaries additionally increases the resistivity. Conventional theories usually introduce grain boundary scattering as an additional source of defect scattering [5, 6], i.e. the assumption (2) is maintained.

The present authors [3, 7–10] studied the thickness-dependent conductivity of polycrystalline metal films using a modified Fuchs model given in [11]. The existence of a (macroscopic) surface roughness (i.e. a fluctuating film cross section) was taken into account. With a four-parameter fit (ρ_∞, l_∞, p and the surface roughness H) the thickness-dependent resistivity was described very well down to small thicknesses. Consequently the determination of the MFP was possible without stressing the assumption (2). Typical accuracies of the fitting parameters are

$$\Delta\sigma_\infty/\sigma_\infty = 0.03 \qquad \Delta l_\infty/l_\infty = 0.15 \qquad \Delta p/p = 0.4 \qquad \Delta H/H = 0.03.$$

The main result of a great number of experiments was surprising. The product $\rho_\infty l_\infty$ (corresponding to a very thick polycrystalline film) depends on the grain boundary scattering of the CEs. This result contradicts conventional theories of the electrical conduction in polycrystalline metals and has been intensively discussed [12]. The main criticism concerned the reliability of the model in [11] for very thin films. The procedure proposed in [3, 7–10] may include changes in film structure (grain size and roughness) during film growth, leading to wrong results for the MFP.

The determination of the MFP from the thickness-dependent resistivity would be improved if

(i) no *a priori* assumptions concerning the product $\rho_\infty l_\infty$ are made and
(ii) the experiments are performed at larger film thicknesses in order to avoid thickness-dependent effects of surface roughness or film structure.

With regard to these demands we present two new, very simple approaches for determining the MFP in polycrystalline films.

(i) In *method 1*, during coating of the metal surface with suitable adatoms, only the surface scattering of CEs increases whereas the film itself remains unchanged. From the maximum relative increase in the resistivity of two identical films, coated at two different temperatures, surface and volume effects can be separated. The MFP can thus be determined very simply and without the uncertainties mentioned before.

(ii) *Method 2* combines the observed thickness dependence of the resistivity in the thickness range of the Sondheimer approximation (equation (1*a*)) with the increase in the resistivity during the subsequent coating at the final thickness. This approach increases the reliability of the obtained results.

2. Experimental methods and results

2.1. *Coating experiments at different temperatures*

Copper films of various thicknesses (10–100 nm) were evaporated in UHV (2×10^{-10} mbar) onto Corning glass substrates held at 300 K. The evaporation parameters (rate (0.1 nm s^{-1}) and substrate temperature) were carefully controlled in order to obtain reproducible film properties. During evaporation the conductivity and the film thickness were accurately monitored as reported elsewhere [3].

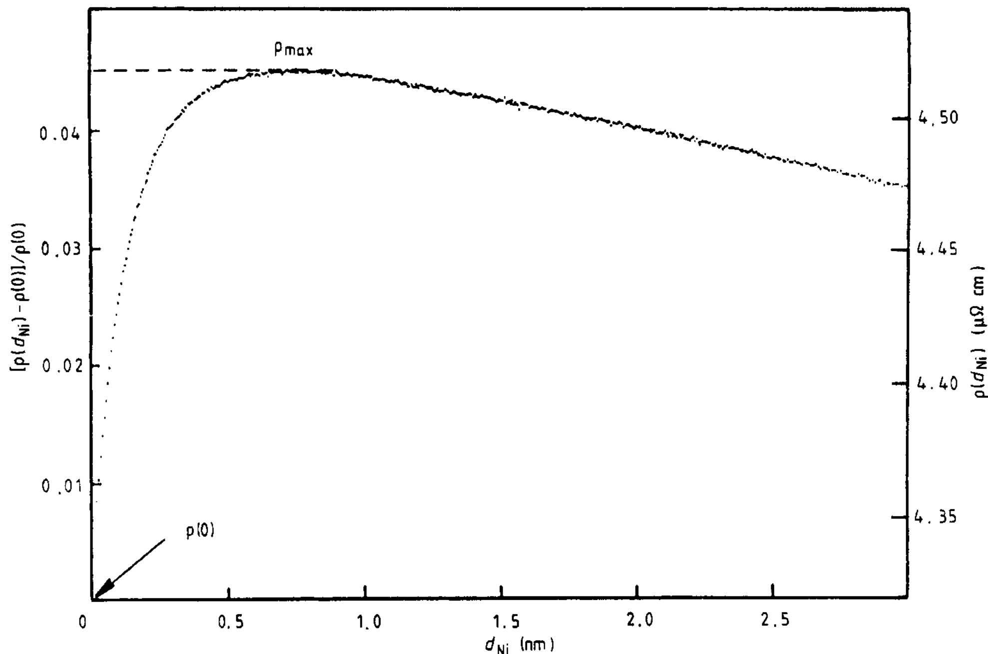

Figure 1. Change in resistivity of a copper film (d = 46.2 nm) during coating with nickel atoms.

After evaporation, one half of the films was slowly coated with nickel atoms (rate, 0.001 nm s^{-1}). The other half was immediately cooled to 80 K (at 6 K min^{-1}) and also coated with nickel or aluminium atoms. A typical example of the resulting increase in the resistivity is given in figure 1.

First the resistivity steeply increases. Every adsorbed atom acts as additional scattering centre. With further coating the probability that adatoms arrive on a free surface site decreases. Consequently the increase in resistivity is not proportional to the number of adsorbed atoms. At the maximum all possible surface sites are occupied [13]. Because of the statistical coating, the mean thickness of the overlayer film at the resistivity maximum is larger than a monolayer, i.e. it is about 0.8 nm for Cu films coated with Ni. This statistical growth mechanism has been additionally confirmed by monitoring the work function of Cu films during coating with Ni atoms [14].

As soon as the overlayer forms a continuous film, the resistance slowly decreases. In this paper, we discuss only the maximal increase $\Delta\rho_{max}/\rho(0)$ in resistivity (see figure 1). The evaluation of the MFP and specularity from these coating experiments can be done using the following assumptions.

(i) During coating with nickel (aluminium) atoms only the surface scattering (specularity) is changed. The volume properties (l_∞ and ρ_∞) are not influenced, i.e. interdiffusion effects are excluded.

(ii) At the maximum resistance all CEs are scattered diffusely on the upper surface.

(iii) Before coating, the specularities at both film surfaces are equal.

Assumptions (i) and (ii) have often been used by other workers [4, 15] in order to explain the enhancement of the resistivity of a metallic thin film during coating with

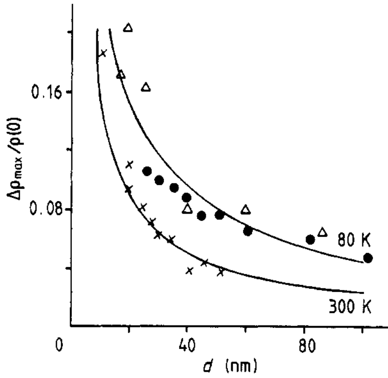

Figure 2. Maximum relative change in resistivity during coating with nickel and aluminium atoms as a function of copper film thickness and substrate temperature: ×, coating with nickel atoms at 300 K; ●, coating with nickel atoms at 80 K; △, coating with aluminium atoms at 80 K; ———, best fit of equations (4) and (6).

adatoms. Because each surface contributes the value $p/2$ in the Fuchs formula [16], the maximal change in specularity during the coating is given by

$$\Delta p_{\max} = -p/2.$$

Consequently

$$\Delta\rho_{\max} = \rho(p/2) - \rho(0) = \tfrac{3}{16}(\rho_\infty l_\infty p/d) \tag{3}$$

where $\rho(0)$ and $\rho(p/2)$ can be expressed by equation (1*a*).

(iv) The specularity is independent of the substrate temperature, as proved in [15].

(v) Decreasing the temperature to 80 K increases the MFP. The temperature dependence of the MFP can be calculated with the Matthiessen rule [17] and the Bloch–Grüneisen equation [18]:

$$\begin{aligned}1/l_\infty\,(80\,\mathrm{K}) &= 1/l_\mathrm{b}\,(80\,\mathrm{K}) + 1/l_\mathrm{s} = 1/l_\mathrm{b}\,(80\,\mathrm{K}) + 1/l_\infty\,(300\,\mathrm{K}) - 1/l_\mathrm{b}\,(300\,\mathrm{K})\\ &= 1/l_\infty\,(300\,\mathrm{K}) - 21.8\times10^{-3}\,\mathrm{nm}^{-1}\end{aligned} \tag{4}$$

where l_b (80 K) = 257 nm is the phonon scattering length at 80 K, l_b (300 K) = 39 nm is the phonon scattering length at 300 K [18] and l_s is the scattering length due to crystal imperfections (temperature independent).

Now the maximal increase in the resistivity can be easily calculated using equation (1*a*):

$$[\beta(d)]_{T=\mathrm{constant}} = \Delta\rho_{\max}/\rho(0) = 3l_\infty p/2[8d + 3l_\infty(1-p)]. \tag{5}$$

As the product $l_\infty p$ is different at different temperatures, $\Delta\rho_{\max}/\rho(0)$ should be larger at 80 K than at 300 K. The ratio $\beta(80\,\mathrm{K})/\beta(300\,\mathrm{K})$ is given by

$$\begin{aligned}\beta(80\,\mathrm{K})/\beta(300\,\mathrm{K}) &= [l_\infty(80\,\mathrm{K})/l_\infty(300\,\mathrm{K})]\\ &\quad\times\{[8d + 3l_\infty(300\,\mathrm{K})(1-p)]/[8d + 3l_\infty(80\,\mathrm{K})(1-p)]\}.\end{aligned} \tag{6}$$

The observed values for copper films coated with nickel and aluminium atoms are given in figure 2 as a function of the film thickness. The change in the resistivity clearly increases at lower temperatures. This shows that this increase is an authentic MFP effect and proves the absence of effects induced by inter-diffusion. The influence of inter-diffusion has been observed only during the coating of Cu films held at 400 K. In this case the resistivity shows no transition through a maximum but permanently increases with increasing thickness of the coating layer.

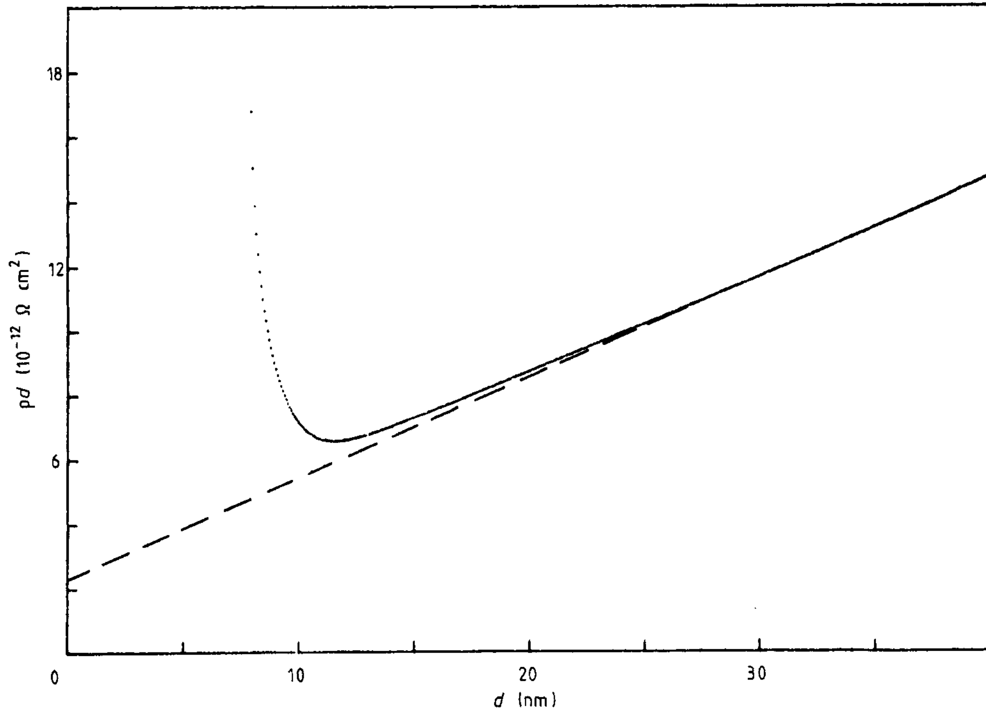

Figure 3. Thickness-dependent resistivity (ρd against d) for copper evaporated at 300 K onto glass substrates.

Finally equations (4) and (6) allow evaluation of the MFP and specularity. The best fit of (4) and (6) to the data, given in figure 2, leads to

$$l_\infty(300\ \mathrm{K}) = 24 \pm 4\ \mathrm{nm} \qquad p = 0.42 \pm 0.07.$$

2.2. Combination of the thickness dependence with coating experiments

The combination of two different experiments, i.e. the thickness dependence of resistivity and surface coating (method 2), can be used to improve the reliability of the electrical transport parameters given above.

2.2.1. Thickness dependence of the resistivity at 300 K. For larger thicknesses ($l_\infty > d$) the product $\rho_\infty l_\infty(1-p)$ can be easily determined from plots of ρd against d (see equation (1*a*)) (figure 3). ρ_∞ is given by the slope; the product $\rho_\infty l_\infty(1-p)$ is determined by the axial intercept of this curve. This is still the standard method used for quantitative interpretations of the thickness-dependent resistivity. For copper films evaporated at 300 K we obtained

$$\rho_\infty = 3.3 \pm 0.3\ \mu\Omega\ \mathrm{cm} \qquad l_\infty(1-p) = 16.0 \pm 2.5\ \mathrm{nm}$$

2.2.2. Surface coating. The maximal increase in the resistivity during coating at 300 K is determined by the product $l_\infty p$ (equation (3)).

For various copper film thicknesses we determined that

$$l_\infty p = 10 \pm 3\ \mathrm{nm}.$$

The resulting values for l_∞ as a function of p for both experiments are shown in figure 4.

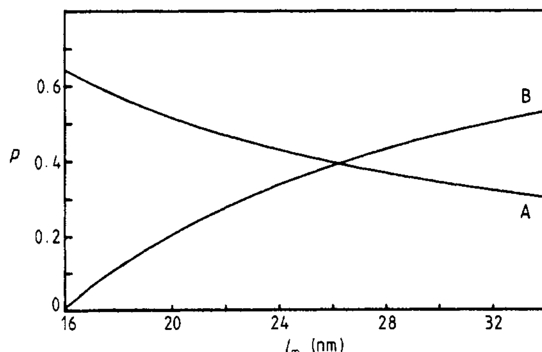

Figure 4. Combination of two independent measurements: curve A, $l_\infty p$ = constant (surface coating); curve B, $l_\infty(1-p)$ = constant (thickness-dependent resistivity).

The intersection between the two functions ($l_\infty p$ and $l_\infty(1-p)$) gives the optimum values of l_∞ and p, i.e.

$$l_\infty = 26 \text{ nm} \qquad p = 0.39.$$

These values are in good agreement with those obtained by coating experiments at different temperatures (method 1).

3. Discussion

The results discussed above clearly show that the MFP in polycrystalline metal films can be simply determined with the methods proposed above. The product $\rho_\infty l_\infty$ related to the polycrystalline thick film, conventionally regarded as a constant of material, was calculated as

$$\rho_b l_b / \rho_\infty l_\infty = 0.76$$

with $\rho_b l_b = 6.57 \times 10^{-12}\ \Omega\ \text{cm}^2$ [18].

This is in very good agreement with previous results obtained by computer fitting of the Fuchs–Namba model to the thickness-dependent conductivity [13], where for the same copper films

$$\rho_\infty = 3.3 \pm 0.3\ \mu\Omega\ \text{cm} \qquad p = 0.41 \pm 0.07$$

$$l_\infty = 26 \pm 2 \text{ nm} \qquad H = 3.8 \pm 0.5 \text{ nm}$$

has been found.

The mean crystallite size of the copper films discussed in this paper determined by transmission electron microscopy is

$$D = 24 \pm 3 \text{ nm}$$

and consequently

$$\rho_b l_b / \rho_\infty l_\infty = 0.76 \qquad \text{for } l_\infty \simeq D.$$

Three different methods therefore proved the following.

(i) The conventional methods for determining the MFP of CEs, using the product $\rho_\infty l_\infty$ as a material constant cannot be applied to polycrystalline metals where the MFP $\geqslant D$.

(ii) Grain boundary scattering cannot be explained by a reduced intrinsic MFP. Therefore the influence of grain boundary scattering on the resistivity has to be discussed in a different way [19].

4. Conclusion

The MFP of thin polycrystalline metal films can be accurately and easily determined from surface coating experiments at two different substrate temperatures. The reliability of the obtained values of l_∞ and p can be increased if two combined methods (surface coating experiments and thickness-dependent resistivity) are considered. Additionally the results show that grain boundary scattering cannot be explained with a reduced intrinsic MFP as required in the conventional models (first-order perturbation).

Acknowledgments

The authors are indebted to Siemens AG, Munich, for support and to G Reiss for carefully reading the manuscript.

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