Temperature dependence of resistivity in thin metallic film

Group meeting (07/02/2021)
Bharat Giri

Resistivity

- Resistivity depends on collision rates of conduction electron with phonons and lattice imperfections
- Matthienssen's rule: $\rho = \rho_t + \rho_i$

First term ρ_t: scattering from phonons

Temperature dependent (solely depends on electron phonon interaction)

Second term ρ_i: scattering from lattice imperfection

independent of temp → residual resistivity by extrapolating to 0K

- High temp: linear Low temp: $AT^2 + BT^5$

- Matthiessen's rule not valid for thin film:

lattice imperfection includes surface and grain boundaries which are temperature dependent-> mean free path

- Temperature dependence of resistivity:

Independent of film thickness

Experimental result: graph alongside

Only affects residual resistivity

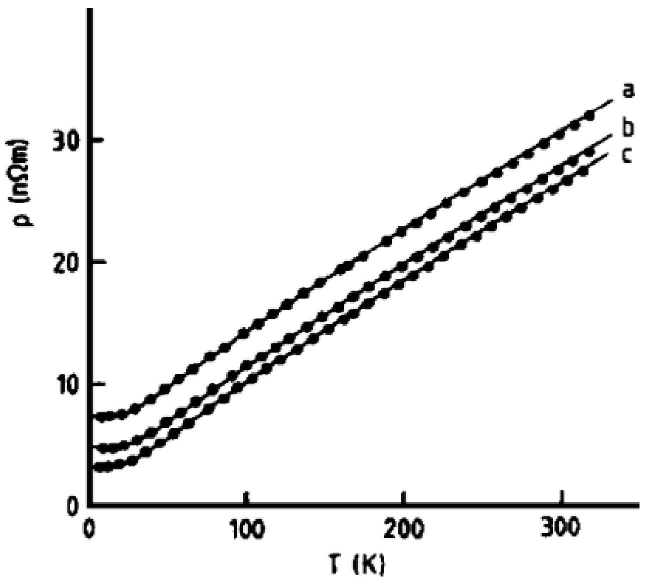


FIG. 1. Resistivity ρ vs temperature T for three annealed gold films of different thicknesses: 18.0 nm (a), 34.6 nm (b), and 82.1 nm (c). From de Vries [Ref. 5].

J. W. C. de Vries, Thin Solid Films 150,201

Mayadas and Shaztkes model(MS model):

Why MS model?
Although there are other more sophisticated quantum approach this model combine the influence of surface and grain boundaries. Thus, not applicable for polycrystalline crystal

Assumptions:
- Semiclassical transport between grain boundaries
- Grain boundaries are perpendicular to transport
- Grain boundaries are translation-invariant along the boundary
- Transmission can be characterized by parameter reflection coefficient(R)
- All boundaries in a sample are identical

Temperature dependence comes with dependence of mean free path.

This dependence doesn't have simple form. Difficult to analyze different factor and separate contribution of surface and grain boundaries

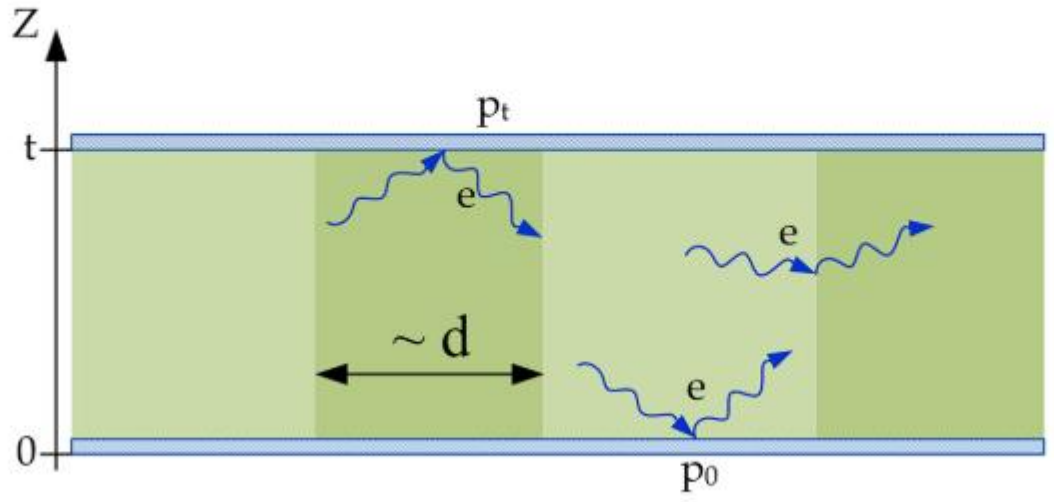


FIG. 1. Schematics of the boundary conditions invented by Fuchs and Sondheimer used by Mayadas and Shatzkes, describing the reflectivity p_0 of the rough surface located at $z = 0$, and the reflectivity p_t of the rough surface located at $z = t$. The shaded areas represent grains exhibiting a different crystalline orientation, separated by an average distance d. The arrows portray the delta function potential representing grain boundaries.

d: (separation distance) shadowed color shows grains exhibiting a different crystalline orientation
p: Specularity parameter(1 for perfectly smooth value or totally specular surface)

Result from MS model:

$$\frac{\rho_B}{\rho_f} = f(\alpha) - g(k,p,\alpha)$$

Where,

$$f(\alpha) = 1 - \frac{3}{2}\alpha + 3\alpha^2 - 3\alpha^3 \ln\left(1 + \frac{1}{\alpha}\right)$$

$$g(k,p,\alpha) = \frac{6(1-p)}{\pi k}\int_0^{\frac{\pi}{2}} d\phi \int_0^{\frac{\pi}{2}} \frac{\cos\theta\, \sin^3\theta \cos^2\phi}{H^2} \frac{1 - e^{-\frac{kH}{\cos\theta}}}{1 - pe^{-\frac{kH}{\cos\theta}}} d\theta$$

$\alpha = \frac{\lambda_B}{d}\frac{R}{1-R}$

$d \rightarrow$ *Diameter of grain,*

$R \rightarrow$ *grain boundary reflection coefficient*

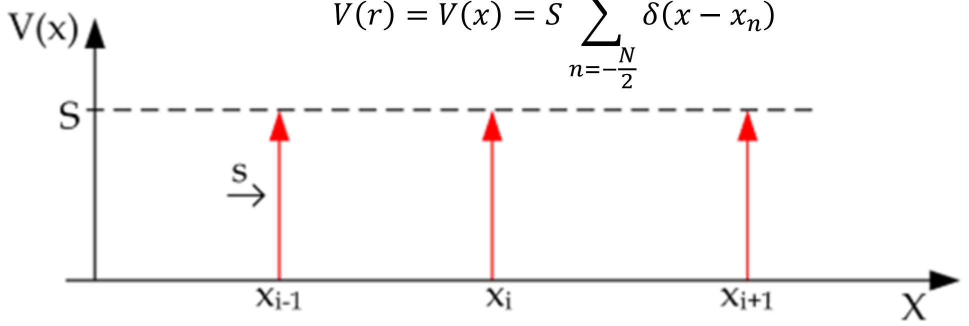
$\lambda_B \rightarrow$ *mean free path*

$p \rightarrow$ *specularity parameter of surface*

$k \rightarrow$ *ratio between the film thickness t and mean free path* $\left(\frac{t}{\lambda_B}\right)$

$H = 1 + \frac{\alpha}{\cos\phi\, \sin\theta}$

$$V(r) = V(x) = S\sum_{n=-\frac{N}{2}}^{\frac{N}{2}} \delta(x - x_n)$$



$$v_z \frac{\partial f_1}{\partial z} + qEv_x \frac{\partial f_0}{\partial \varepsilon} = \frac{f_1}{\tau} + \int P\left(\vec{k}', \vec{k}\right)\left[f_1\left(\vec{k}'\right) - f_1\left(\vec{k}\right)\right] d^3k'$$

$$f_1(k) = f(k) - f_0(k)$$

$P(k,k') =$ *Transition probability* $(k' \rightarrow k)$

For $N \rightarrow \infty$ *and equally spaced potential*
$\rightarrow$ *Kronig Penney potential*

$$\frac{\rho_B}{\rho_f} = f(\alpha(T)) - g(k(T), p, \alpha(T))$$

Free electron approximation:

$$\lambda_B(T)\rho_B(T) = constant$$

T dependence of film resistivity:

$$\frac{d\rho_f}{dT} = \frac{\rho_f}{\rho_B}\frac{d\rho_B}{dT} - \frac{\rho_f^2}{\rho_B}\frac{df}{dT} + \frac{\rho_f^2}{\rho_B}\frac{dg}{dT}$$

1.

$$\frac{dg}{dT} = \frac{6(1-p)}{\pi}\int_0^{\frac{\pi}{2}} d\phi \int_0^{\frac{\pi}{2}} cos\theta \; sin^3\theta \cos^2\phi \frac{d}{dT}\left(\frac{1}{kH^2}\frac{1-e^{-\frac{kH}{cos\theta}}}{1-pe^{-\frac{kH}{cos\theta}}}\right) d\theta$$

$$\frac{d}{dT}\left(\frac{1}{kH^2}\frac{1-e^{-\frac{kH}{cos\theta}}}{1-pe^{-\frac{kH}{cos\theta}}}\right) = \left(\frac{1}{kH^2}\frac{1-e^{-\frac{kH}{cos\theta}}}{1-pe^{-\frac{kH}{cos\theta}}}\right)(1+n)\frac{1}{\rho_B}\frac{d\rho_B}{dT}$$

$$Where, n = \frac{k(1-p)e^{-\frac{kH}{cos\theta}}}{cos\theta\left(1-pe^{-\frac{kH}{cos\theta}}\right)\left(1-e^{-\frac{kH}{cos\theta}}\right)} - \frac{2}{H}$$

$$\Rightarrow \frac{dg}{dT} = \frac{1}{\rho_B}\frac{d\rho_B}{dT} q$$

Where,

$$q = \frac{6(1-p)}{\pi k}\int_0^{\frac{\pi}{2}} d\phi \int_0^{\frac{\pi}{2}} cos\theta \; sin^3\theta \cos^2\phi \left(\frac{1}{H^2}\frac{1-e^{-\frac{kH}{cos\theta}}}{1-pe^{-\frac{kH}{cos\theta}}}\right)(n+1) d\theta$$

$$H = 1 + \frac{\alpha}{cos\phi \; sin\theta} \qquad \alpha = \frac{\lambda_B}{d}\frac{R}{1-R}$$

$$x = \frac{kH}{\cos\theta} \qquad \lambda_B(T)\rho_B(T) = constant$$

$$\frac{d}{dT}\left[\ln\left(\frac{1}{kH^2}\frac{1-e^{-x}}{1-pe^{-x}}\right)\right] = -\frac{1}{k}\frac{dk}{dT} - \frac{2}{H}\frac{dH}{dT} + \frac{e^{-x}(1-p)}{(1-e^{-x})(1-pe^{-x})}\frac{dx}{dT}.$$

$$\frac{dH}{dT} = \frac{1}{\cos\phi\sin\theta}\frac{d\alpha}{dT} = -\frac{1}{\cos\phi\sin\theta}\frac{\alpha}{\rho_B}\frac{d\rho_B}{dT} = (1-H)\frac{1}{\rho_B}\frac{d\rho_B}{dT}.$$

$$\frac{dx}{dT} = \frac{1}{\cos\theta}\frac{d(kH)}{dT} = \frac{1}{\cos\theta}\left(k(1-H)\frac{1}{\rho_B}\frac{d\rho_B}{dT} + H\frac{k}{\rho_B}\frac{d\rho_B}{dT}\right) = \frac{1}{\cos\theta}\frac{k}{\rho_B}\frac{d\rho_B}{dT}.$$

2.

$$\frac{df}{dT}=\frac{df}{d\alpha}\frac{d\alpha}{dT}=\frac{df}{d\alpha}\frac{1}{D}\frac{R}{1-R}\frac{d\lambda_B}{dT}=\frac{df}{d\alpha}\frac{\alpha}{\lambda_B}\left(-\frac{c}{\rho_B^2}\frac{d\rho_B}{dT}\right)=-\frac{df}{d\alpha}\frac{\alpha}{\rho_B}\frac{d\rho_B}{dT}$$

$$\frac{d\rho_f}{dT}=\left(\frac{\rho_B}{\rho_f}+\alpha\frac{df}{d\alpha}+q\right)\left(\frac{\rho_f}{\rho_B}\right)^2\frac{d\rho_B}{dT}$$

Using 1 and 2,

$$\frac{\rho_f'}{\rho_B'}=\frac{\left(f-g+\alpha\frac{df}{d\alpha}+q\right)}{(f-g)^2}$$

$$=\frac{\left(f+\alpha\frac{df}{d\alpha}+s\right)}{(f-g)^2}$$

$$\frac{\rho_B(T)}{\rho_f(T)}=f[\alpha(T)]-g[k(T),p,\alpha(T)].$$

s = q – g

$$g=\int_0^{\pi/2}d\phi\int_0^{\pi/2}m\,d\theta,$$

$$q=\int_0^{\pi/2}d\phi\int_0^{\pi/2}m(n+1)\,d\theta$$

$$m=\frac{6(1-p)}{\pi k}\frac{\cos\theta\sin^3\theta\cos^2\phi}{H^2}\frac{1-e^{-kH/\cos\theta}}{1-pe^{-kH/\cos\theta}}$$

Pure grain boundary effect with α (wide range)

$$\alpha = \frac{\lambda_B}{d}\frac{R}{1-R}$$

$\alpha \rightarrow$ Temperature dependent through λ_B

Totally specular surfaces:
p = 1 and q = g = 0

$$\frac{\rho_f'}{\rho_B'} = \frac{\left(f + \alpha\frac{df}{d\alpha}\right)}{(f)^2}$$

- Value of ratio close to 1. Small dependence of temperature dependent part of resistivity on thickness

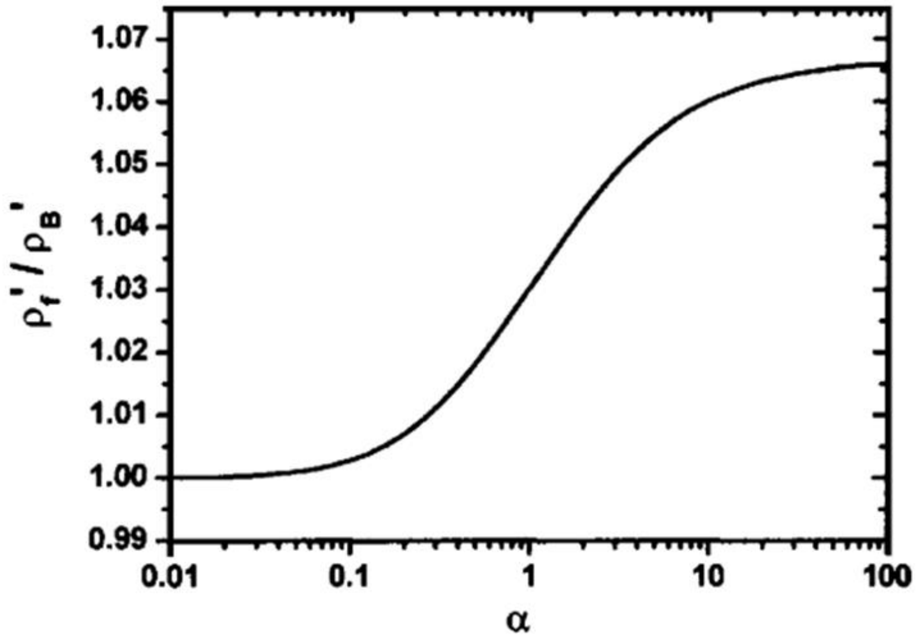


FIG. 2. The dependence on α of the ratio of the film derivative (with respect to temperature) to the bulk derivative for a pure grain boundary effect with no surface effect ($p=1$). There is almost no thickness dependence of the temperature-dependent part of the resistivity.

Effect of surface specularity and film thickness on temperature dependent resistivity:

Dependence of the derivative ratio on k for different specularity.

No grain boundaries effect(R=0):

$$\alpha = 0 \ \rightarrow f = 1$$

$$\frac{\rho_f'}{\rho_B'} = \frac{(1+s)}{(1-g)^2}$$

- For non specular surface, clear deviation is seen at low temperature.
- When k =0.4(Mean free path is approximately twice of the film thickness), deviation is about 10% in case of maximal surface effect.
- An important implication is to extract the specularity parameter of single crystalline from the measurement.

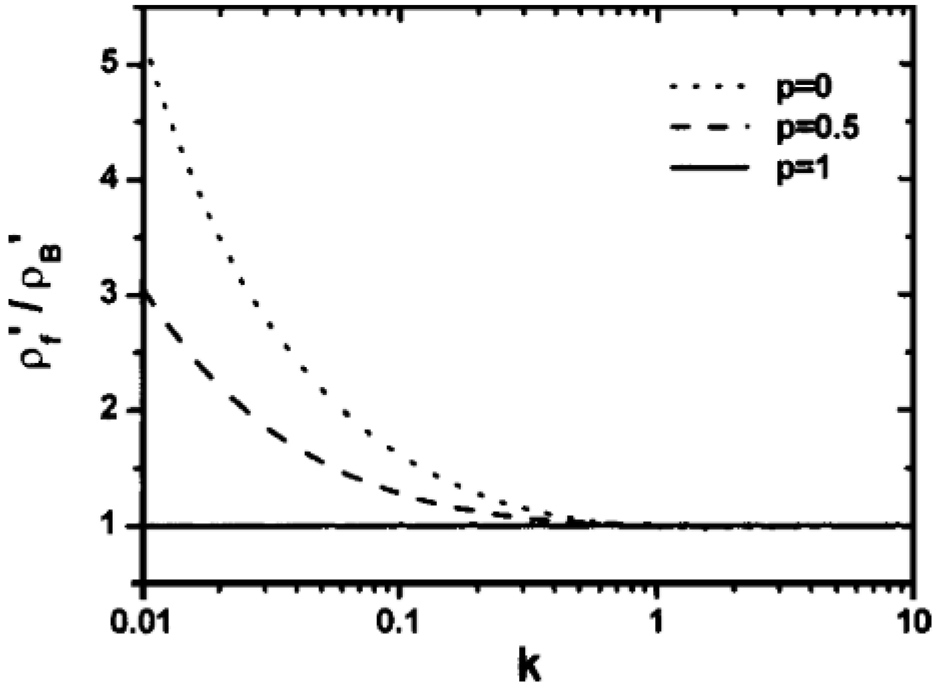


FIG. 3. The dependence on k of the ratio of the film derivative to the bulk derivative for a pure surface effect with no grain boundary effect (α=0). A clear deviation from bulk behavior is identified for nonspecular surfaces.

Dependence of derivative ratio on k at specific p.

- We use the common approximation that the mean grain size is equal to the film thickness.

$$k\alpha = \frac{t}{\lambda_B}\frac{\lambda_B}{D}\frac{R}{1-R} = \frac{t}{D}\frac{R}{1-R} \cong \frac{R}{1-R}$$

- kα depends only on the grain boundary reflection coefficient
- One can see, for example, that for R=0.3 and p=0 no substantial deviation from bulk is identified even for k values as low as 0.01 at low temperature. For this set of values, both grain boundary scattering and surface scattering are important, and none of them can be neglected.

- It means that a weak thickness dependence of the temperature-dependent part of the resistivity does not necessarily mean that grain boundary scattering is dominant.
- The combined influence of grain boundaries and surfaces can sometimes be an appropriate alternative explanation.

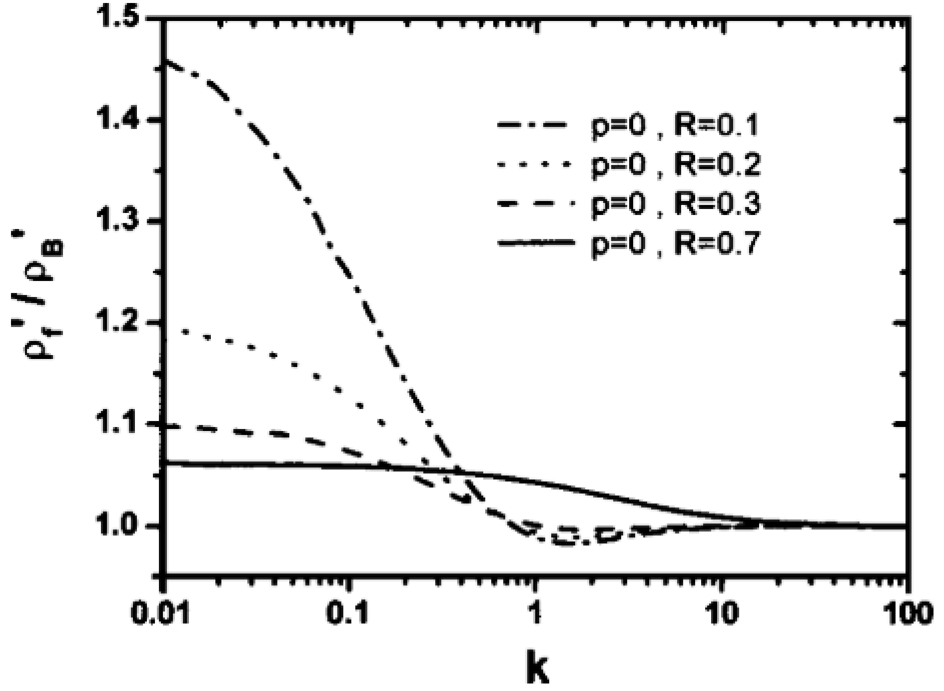


FIG. 4. The dependence on k of the ratio of the film derivative to the bul derivative for maximal surface effect (p=0) and different grain boundar reflection coefficients (R values). Both grain boundaries and surfaces influ ence the temperature-dependent part of the resistivity.

Comparison of resistivity at R=0.3 and p=0 with calculated resultant resistivity to that obtained for other R and p pairs

- Presents the dependence of the resistivity ratio $\frac{\rho_f(R,p)}{\rho_f(0.3,0)}$
- Comparison of the reference case with the results for the pure grain boundary effect R=0.3, p=1 and for the pure surface effect R=0,p=0
- An important conclusion can be deduced from the results obtained for the pure grain boundary effect p=1 with R =0.425.

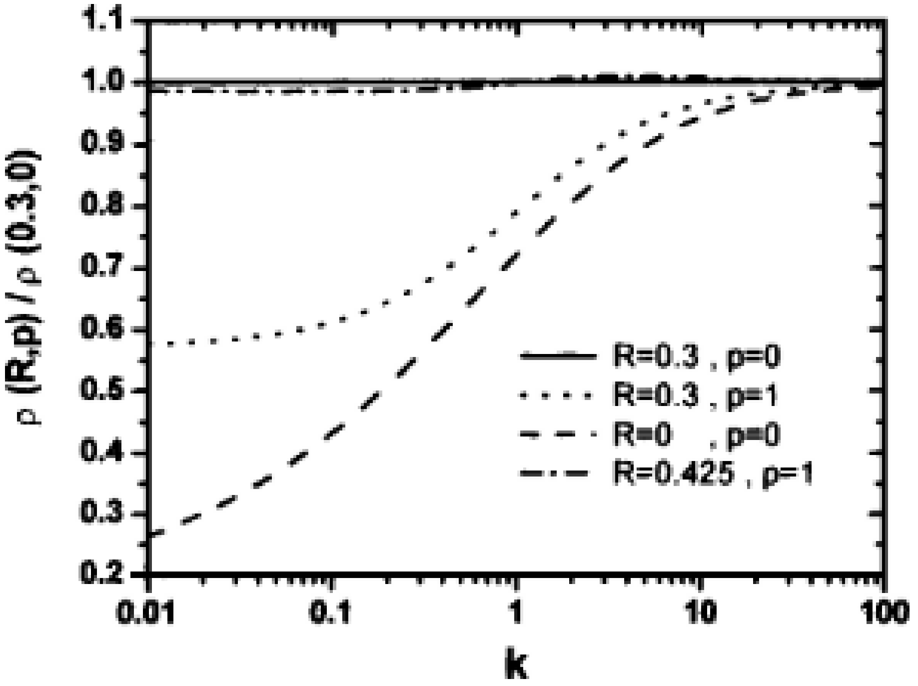


FIG. 5. The dependence of the resistivity ratio $\rho_f(R,p)/\rho_f(0.3,0)$ on k for different R and p values. $\rho_f(R,p)$ is the resistivity of a film with a grain boundary coefficient R and a specularity parameter p. For the reference case of $R=0.3$ and $p=0$ neither grain boundaries nor surfaces can be neglected. Almost identical resistivity-temperature dependence is expected for both this case and the case of a pure grain boundary effect ($p=1$) with $R=0.425$.

Summary

- The analytical expressions developed enable us to systematically study the expected dependence of resistivity on temperature for thin metal films. Using these expressions, the effect of the different influencing factors can be quantitatively studied.
- It was shown that the combined influence of grain boundaries and surfaces is of great importance in analysis of experimental results.
- Previously suggested interpretations of a dominant grain boundary mechanism are not necessarily correct. Conclusions regarding specific values can be tested using the proposed model.

Thank you!