

Classical theory of electron grain boundary and surfaces scattering

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Result used in previous presentation:

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

Where,

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

$g(k, p, \alpha)$

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

$d \rightarrow$ Average separation of grain boundaries (GB)

$D \rightarrow$ Diameter of grain,

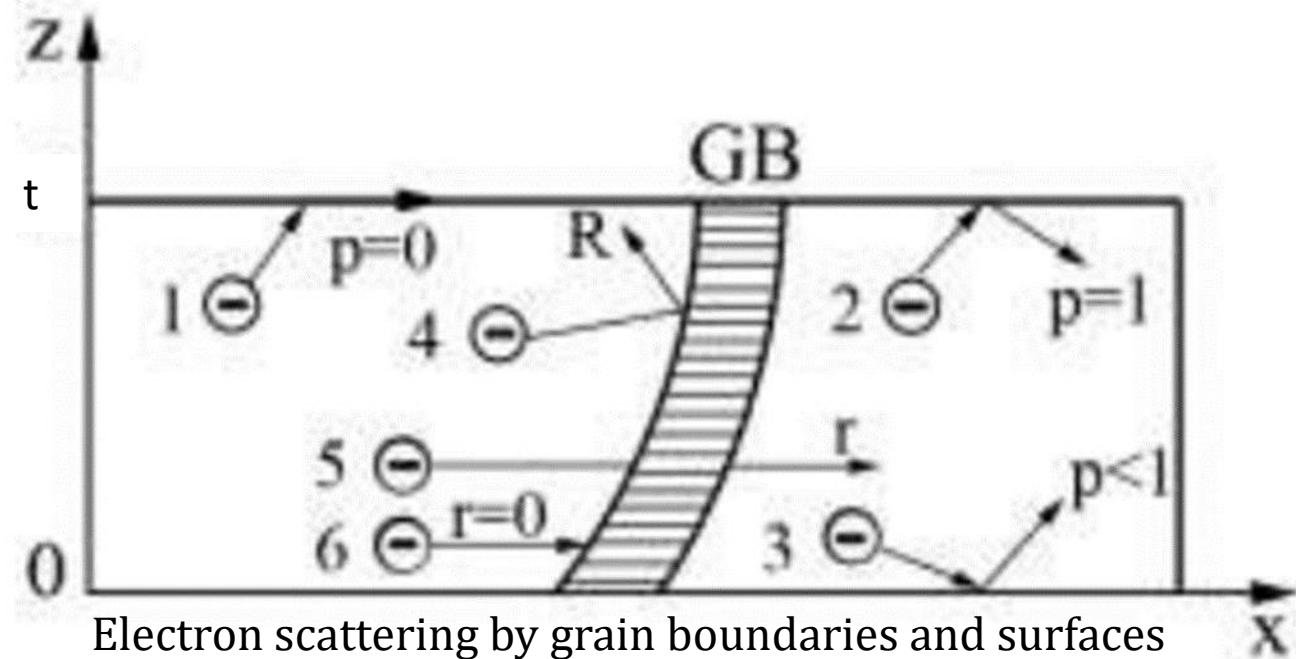
$R \rightarrow$ grain boundary reflection coefficient

$l_0 \rightarrow$ mean free path

$p \rightarrow$ specularity parameter of surface

$$k \rightarrow \frac{t}{l_0} \quad \alpha = \frac{l_0}{d} \frac{R}{1-R}$$

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



Electron scattering by grain boundaries and surfaces

$$p = \begin{cases} 0 & \text{entirely diffuse} \quad (1) \\ 1 & \text{entirely specular scattering} \quad (2) \end{cases}$$

4, 5, 6 : transition from GB

Overview:

Scattering by grain boundaries:	MS model
Scattering by surfaces:	FS model
Background scattering: (point defects and phonons)	Bulk

Boltzmann transport equation:

$$\left(\frac{\partial f^1}{\partial t} \right)_{coll} + \left(\frac{\partial f^1}{\partial t} \right)_{drift} = \frac{\partial f^1}{\partial t}$$

Linearized BTE,

$$\left(\frac{\partial f^1}{\partial t} \right)_{coll} = \frac{\partial f^1}{\partial t} + \frac{e}{\hbar c} (\mathbf{v} \times \cancel{\mathbf{B}}) \nabla_{\mathbf{k}} f^1 + v(k) \cdot \left(\cancel{\nabla_r T} \frac{\partial f^0}{\partial t} + e \boldsymbol{\epsilon} \frac{\partial f^0}{\partial \epsilon} \right)$$

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

$$f_0 = \frac{1}{e^{\frac{\epsilon_i - \mu}{k_B T}} + 1}$$

MS Model:

ρ_g : Scattering from series of partially reflecting grain boundaries and background scattering from phonons and defects (Bulk)

Assumption:

Grain boundaries normal to surface (randomly spaced)

Grain boundary potential is short ranged and smooth (delta potential)

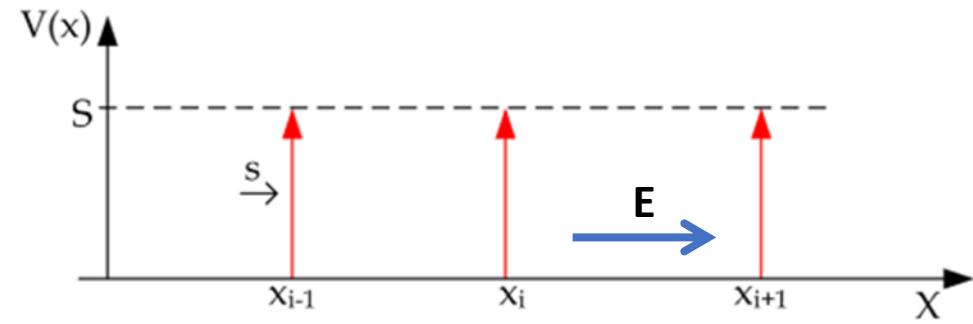
BTE

$$e\mathbf{v}(\mathbf{k}) \cdot \mathbf{E} \frac{\partial f^0}{\partial \epsilon} = \left(\frac{\partial f^1}{\partial t} \right)_{coll} = \int P(\mathbf{k}, \mathbf{k}') [\Phi(\mathbf{k}) - \Phi(\mathbf{k}')] d\mathbf{k} + \frac{\Phi(\mathbf{k})}{\tau}$$

Grain boundaries Background

Perturbation theory:

$$\text{Transition probability } (k' \rightarrow k) = P(k, k') = \left(\frac{2\pi}{\hbar} \right) |\langle \mathbf{k}|V|\mathbf{k}' \rangle|^2 \delta(\epsilon_k - \epsilon_{k'})$$



- Potential $S\delta(x - x_n)$
- Average separation: d
- Grain boundary separation follows gaussian distribution:

$$g(x_1, \dots, x_N) = \frac{\exp \left[- \sum_{i=1}^{N-1} \frac{(x_{i+1} - x_i - d)^2}{2s^2} \right]}{L_x (2\pi s^2)^{\frac{N-1}{2}}}$$

Transition probability:

For free electron,

$$\psi_k = \frac{1}{\sqrt{V}} e^{ik \cdot r}$$

Delta function:

$$\delta(x - x_n) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{ik(x-x_n)} dk$$

$$\langle \mathbf{k}|V|\mathbf{k}'\rangle = \left(\frac{S}{L_x}\right) \delta(k_t - k'_t) \sum_{n,n'} e^{i(k_x - k'_x)(x_n - x_{n'})}$$

k_t : component in yz plane

Averaging over gaussian distribution: $\int g(x_i) P(k, k') dx_i$

$$P(\mathbf{k}, \mathbf{k}') = F(|k_x|) \delta(k_t - k'_t) \delta(k_x + k'_x)$$

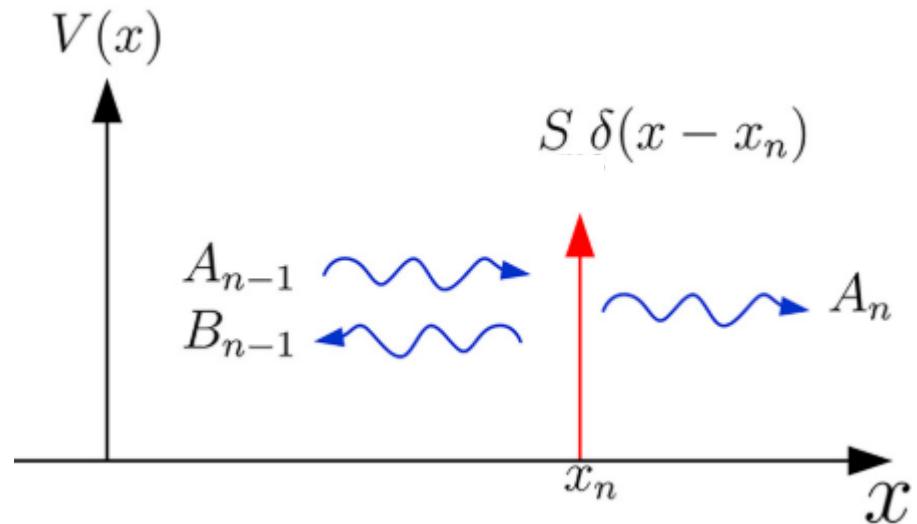
Where,

$$F(|k_x|) = \frac{mS^2}{\hbar^3 d |k_x|} \frac{1 - e^{-4k_x^2 s^2}}{1 + e^{-4k_x^2 s^2} - 2e^{-2k_x^2 s^2} \cos 2k_x d}$$

$$\boxed{\frac{\alpha}{2\tau} \frac{k_F}{|k_x|}}$$

$$\alpha = \frac{2mS^2\tau}{\hbar^3 dk_F} = \frac{l_0}{d} \frac{R}{1-R}$$

Reflection coefficient of single grain boundary



- Boundaries are identical with same R
 - Equally spaced boundaries (Translational symmetry)
- $$\Phi_{n-1}(x) = A_{n-1} e^{ik_x x} + B_{n-1} e^{-ik_x x} \quad \text{for } (n-1)d \leq x \leq nd$$
- $$\Phi_n(x) = A_n e^{ik_x x} + B_n e^{-ik_x x} \quad \text{for } nd \leq x \leq (n+1)d$$
- Boundary conditions:

$$\Phi_{n-1}(x = x_n) = \Phi_n(x = x_n)$$

$$\left(\frac{d\Phi_n(x)}{dx} \right)_{x=x_n} - \left(\frac{d\Phi_{n-1}(x)}{dx} \right)_{x=x_n} - \frac{2m}{\hbar^2} S \Phi(x = x_n) = 0$$

$$R = \left| \frac{B_{n-1}}{A_{n-1}} \right|^2 = \frac{m^2 S^2}{(\hbar^2 k)^2 + m^2 S^2}$$

$$\text{For } B_n=0, S^2 = \left(\frac{\hbar^2 k_x}{m} \right)^2 \frac{R}{1-R}$$

Resistivity by grain boundary scattering:

Boltzmann equation solution:

$$\Phi(k) = \tau^* e E v_x \left(\frac{\partial f_0}{\partial \epsilon} \right) \text{ where } \frac{1}{\tau^*} = \frac{1}{\tau} + 2F(|k_x|)$$

For interplanar spacing(t) \approx Average grain diameter (d) $\Rightarrow k_F^2 s^2 \gg 1$

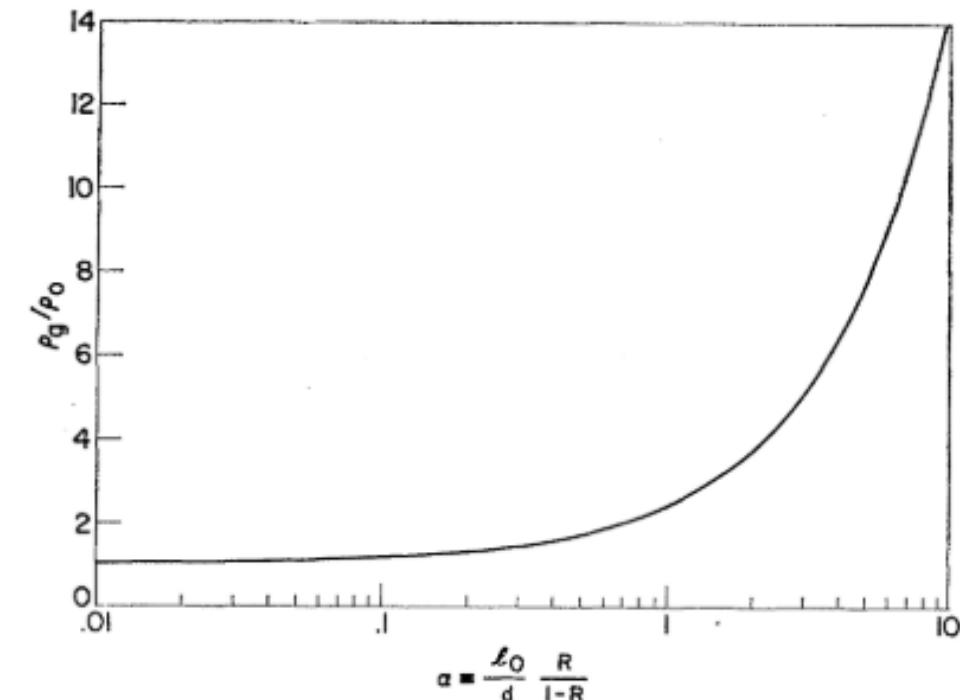
$$\Rightarrow \tau^* = \frac{\tau}{1 + \frac{\alpha k_F}{k_x}}$$

Integrating over Fermi sphere,

$$\sigma_g = \frac{e^2}{4\pi^3} \int \frac{\tau^* v_x^2}{|\nabla_k \epsilon|} dS_F = \frac{3\sigma_0}{2\tau} \int_{-1}^1 \tau^*(q) q^2 dq$$

Where, $q = \cos\theta$, $k_x = k_F q$, $\nabla_k \epsilon = h\nu$ and $\sigma_0 = \frac{ne^2\tau}{m}$

$$\begin{aligned} \frac{\sigma_g}{\sigma_0} &= \frac{3}{2} \int_{-1}^1 \frac{1}{1 + \alpha/q} q^2 dq = 1 - \frac{3}{2}\alpha + 3\alpha^2 - 3\alpha^3 \ln\left(1 + \frac{1}{\alpha}\right) \\ &= f(\alpha) \end{aligned}$$



$\alpha \uparrow \quad \text{as } d \downarrow \text{ or } R \uparrow$

Fuchs and Sondheimer (FS) model

Boltzmann equation:

$$v_z \left(\frac{\partial \Phi}{\partial z} \right) + e \mathbf{v}_x \mathbf{E} \frac{\partial f^0}{\partial \epsilon} = \left(\frac{\partial f^1}{\partial t} \right)_{coll} = \frac{\Phi}{\tau^*}$$

Boundary condition:

$$\Phi^+(v_z, 0) = p\Phi^-(-v_z, 0)$$

$$\Phi^-(-v_z, t) = p\Phi^+(v_z, t)$$

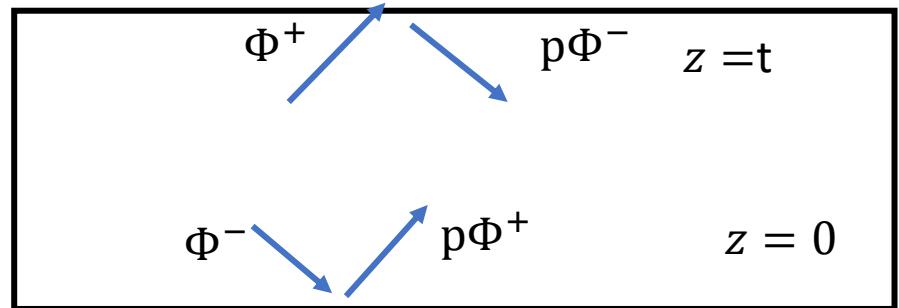
Solution of BTE: $\Phi(v, z) = Ae^{Bz}$ where $B = -1/v_z \tau^*$

$$\Phi^+ = e\tau^* E v_x \frac{\partial f^0}{\partial \epsilon} \left[1 - \frac{(1-p)e^{-\frac{z}{\tau^* v_z}}}{1 - pe^{-\frac{t}{\tau^* v_z}}} \right] \quad \text{for } v_z > 0$$

$$\Phi^- = e\tau^* E v_x \frac{\partial f^0}{\partial \epsilon} \left[1 - \frac{(1-p)e^{-\frac{t-z}{\tau^* v_z}}}{1 - pe^{-\frac{t}{\tau^* v_z}}} \right] \quad \text{for } v_z < 0$$

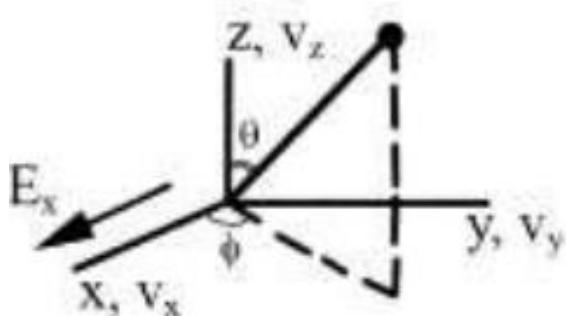
$$J(z) = -\frac{e}{4\pi^3} \left(\frac{m}{\hbar} \right)^3 \int v_x \Phi d\mathbf{v}$$

$$\sigma_f = \frac{1}{Et} \int_0^t J(z) dz$$



Fuchs and Sondheimer Theory:

$$\sigma_f = \frac{e^2}{4\pi^3} \left(\frac{m}{\hbar}\right)^3 \int \tau^* v_x^2 \frac{\partial f^0}{\partial \epsilon} \left[1 - \frac{(1-p)\tau^* v_z \left(1 - e^{-\frac{d}{\tau^* v_z}}\right)}{t \left(1 - pe^{-\frac{d}{\tau^* v_z}}\right)} \right] dv$$



$$v_x = v \cos \phi \sin \theta, \quad v_z = v \cos \theta$$

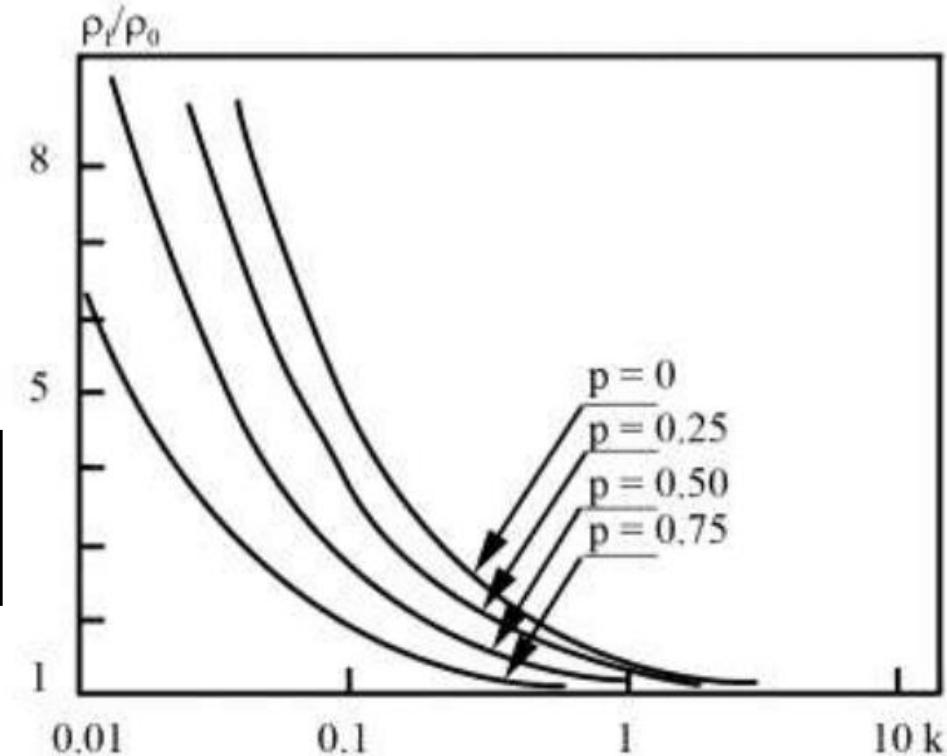
$$\tau^* = \frac{\tau}{1 + \frac{\alpha k_F}{k_x}} = \frac{\tau}{1 + \frac{\alpha}{\cos \phi \sin \theta}} = \frac{\tau}{H}$$

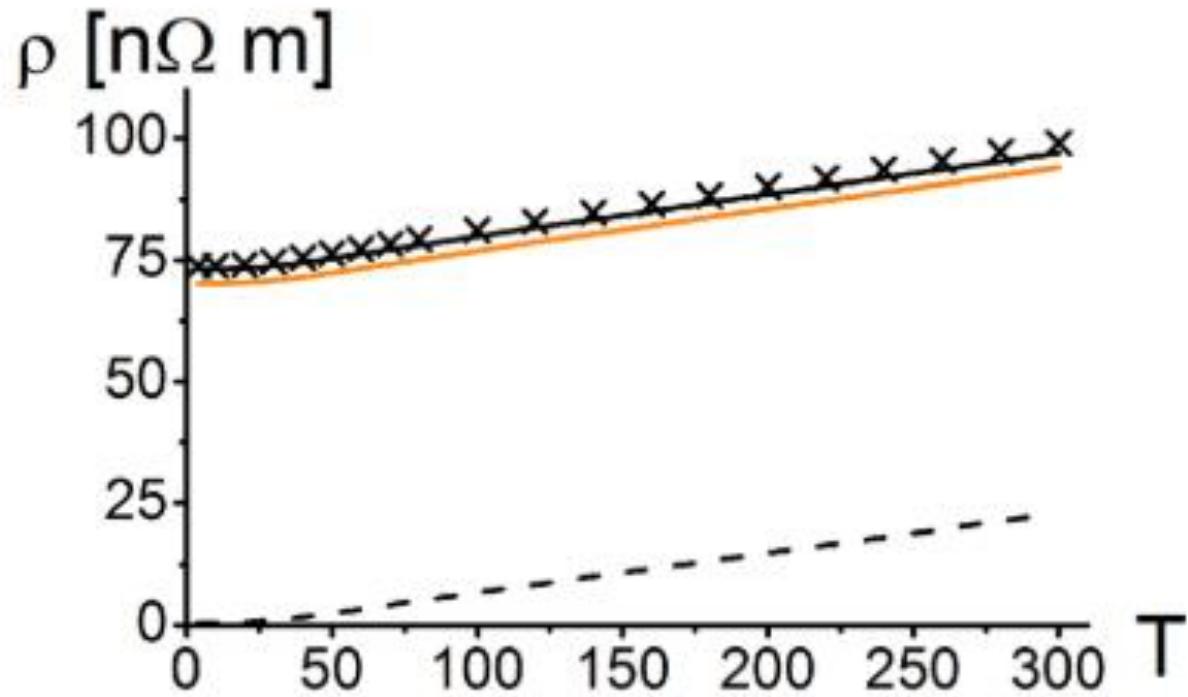
$$dv = dv \sin \theta d\theta d\phi, \quad k = \frac{t}{l_0}, \quad \tau v_F = l_0$$

$$\sigma_f = \left[\sigma_g - \frac{6(1-p)\sigma_0}{\pi k} \int_0^{2\pi} d\phi \int_0^{\frac{\pi}{2}} \frac{\cos \theta \sin^3 \theta \cos^2 \phi}{H^2} \frac{1 - e^{-\frac{kH}{\cos \theta}}}{1 - pe^{-\frac{kH}{\cos \theta}}} d\theta \right]$$

$$\frac{\rho_0}{\rho_f} = \frac{\sigma_f}{\sigma_0} = \left[\frac{\sigma_g}{\sigma_0} - \frac{6(1-p)}{\pi k} \int_0^{2\pi} d\phi \int_0^{\frac{\pi}{2}} \frac{\cos \theta \sin^3 \theta \cos^2 \phi}{H^2} \frac{1 - e^{-\frac{kH}{\cos \theta}}}{1 - pe^{-\frac{kH}{\cos \theta}}} d\theta \right]$$

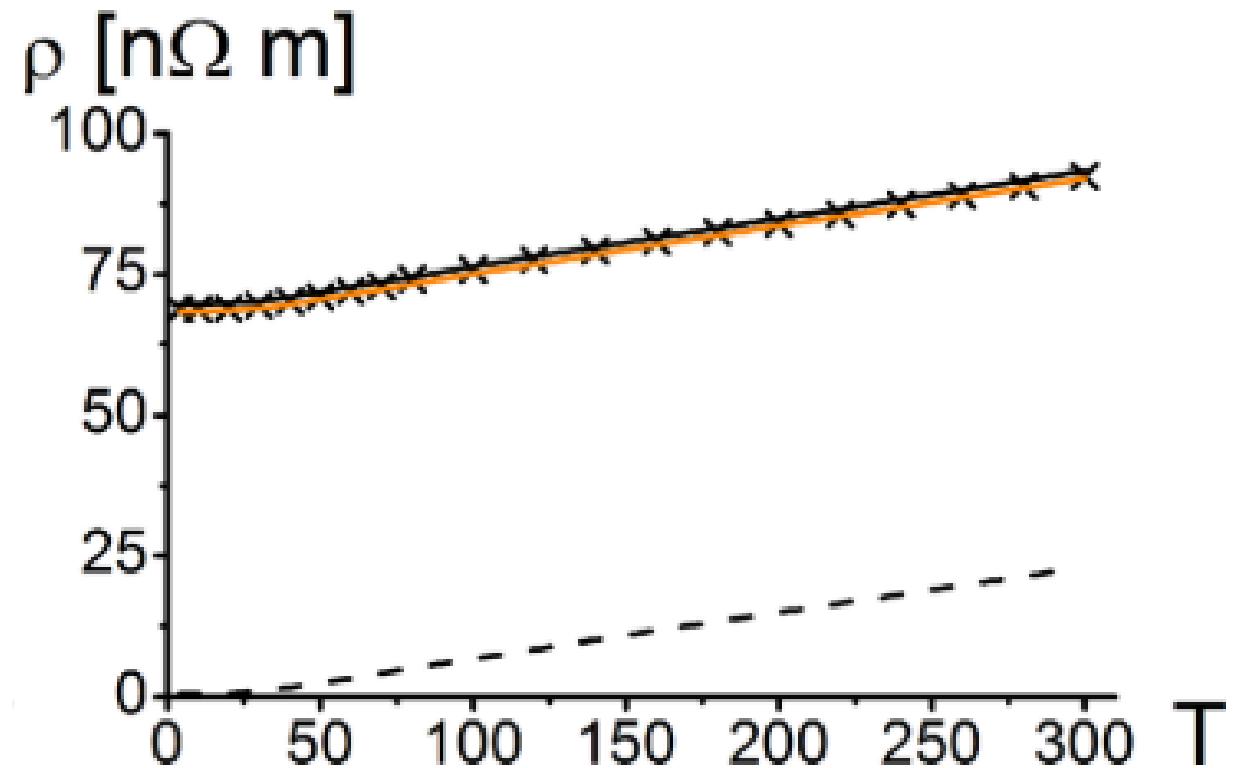
$$\frac{\rho_0}{\rho_f} = f(\alpha) - g(\alpha, p, k)$$





Gold onto mica substrate:
 $t=49\text{nm}$
 $d = 11.1\text{nm}$

Experimental data: Black cross
 Bulk resistivity: Black dash
 $\rho_0 + \text{MS}$: Orange line ($p_0 = p_t = 1$)
 $\rho_0 + \text{MS} + \text{RS(FS)}$: Black line ($p_0 = 0, p_t = 1$)



Gold onto mica substrate:
 $t=109\text{nm}$
 $d = 12.4\text{nm}$

Conceptual difficulties and inconsistencies:

- Inadequate description of the wave functions as plane wave in the presence of grain boundaries.
$$\Phi_k(r) = \exp(ik \cdot r) u_k(r)$$
- For crystalline sample with grain boundaries should include the allowed and forbidden states by GB periodicity.
- Equally spaced grain boundaries