Mechanisms of the Resistivity Size Effect in Metallic Thin Films Computed Using Realistic Tight Binding Models and the Kernel-Polynomial Method

Patrick K. Schelling, Eduardo R. Mucciolo, and William E. Richardson

Department of Physics
University of Central Florida

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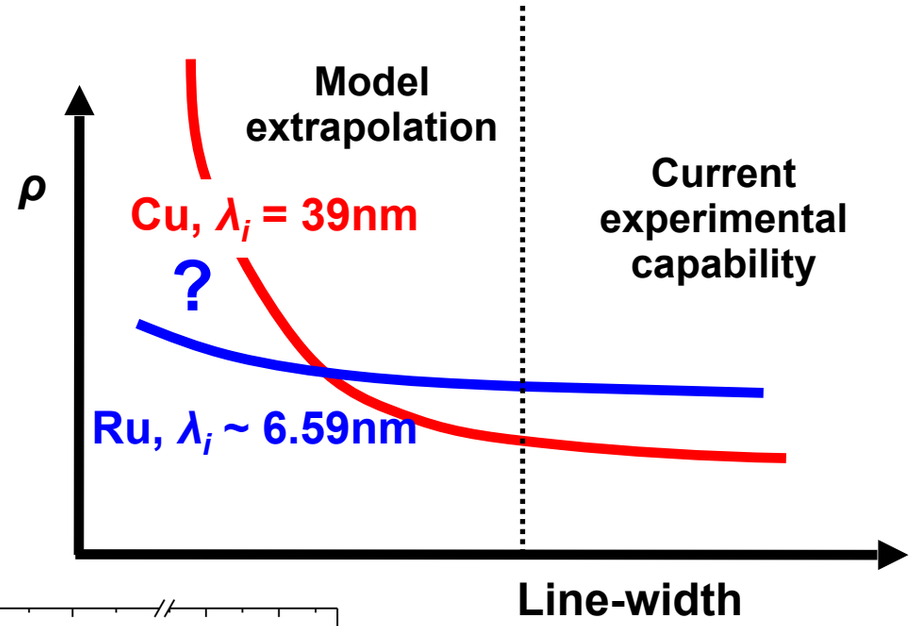
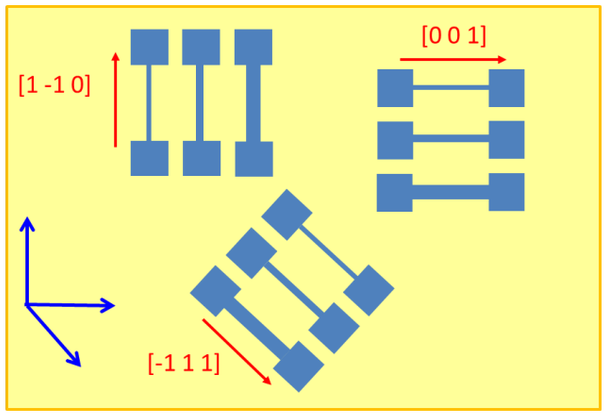
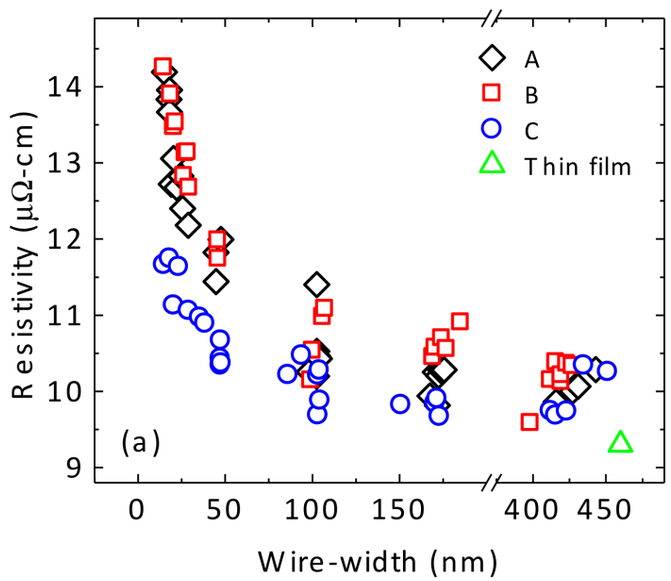
Abstract K66:00007, March 15, 4:12pm

Classical size effect in single-crystalline films and wires

- Electron scattering leads to size-dependent resistivity.
- Possibly metals with shorter bulk mean free paths could be superior to Cu at smaller scales [1].
- Results for W nanowires suggest single-crystal wires exhibit resistivity size effect which depends on crystallographic orientation [2].







[1] Daniel Gall, "Electron mean free path in elemental metals," *J. Appl. Phys.* **119**, 085101 (2016)
[2] D. Choi *et al*, "Crystallographic anisotropy of the resistivity size effect in single crystal tungsten nanowires," *Sci. Rep.* **3**, 2591 (2013)

Origin of surface scattering and early theoretical treatments

- Usually described as “roughness” — but microscopic origin or dominant mechanism not clear.

- Point defects, substrate, surface steps, all cited as possible explanations for the observed effect.

- Simple theoretical models include the Fuchs-Sondheimer model,

$$\rho = \rho_0 \left[1 + \left(\frac{3}{8} \frac{\lambda(1-p)}{d} \right) \right]$$

- Quite often the specularity parameter $p = 0$ is required to fit experimental data [3,4], indicating completely diffusive surface scattering — not clear what produces this strong surface scattering.

[3] E. Milosevic *et al*, “Resistivity size effect in epitaxial Ru(0001) layers,” *J. Appl. Phys.* **124**, 165105 (2018)
[4] S. S. Ezzat *et al*, “Resistivity and surface scattering of (0001) single crystal ruthenium thin films,” *J. Vac. Sci. Tech.* **37**, 031516 (2019)

Orthogonal Tight-binding model for Ru

- The states can be written as a vector, which amounts to a linear combination of orbitals

$$|\psi_\lambda\rangle = \sum_{i,\kappa} c_{\lambda,i\kappa} |\phi_{i\kappa}\rangle \qquad \psi_\lambda(\vec{r}) = \sum_{i,\kappa} c_{\lambda,i\kappa} \phi_{i\kappa}(\vec{r})$$

- The hamiltonian operator connects sites together (hopping)

$$\hat{H} = \sum_{i\kappa} \epsilon_{i\kappa} |\phi_{i,\kappa}\rangle\langle\phi_{i,\kappa}| + \sum_{i\kappa \neq j\kappa'} t_{i\kappa,j\kappa'} |\phi_{i,\kappa}\rangle\langle\phi_{j,\kappa'}|$$

$$H_{i\kappa,j\kappa'} = \delta_{i\kappa,j\kappa'}\epsilon_{i\kappa} + (1 - \delta_{i\kappa,j\kappa'})t_{i\kappa,j\kappa'}$$

Onsite energy

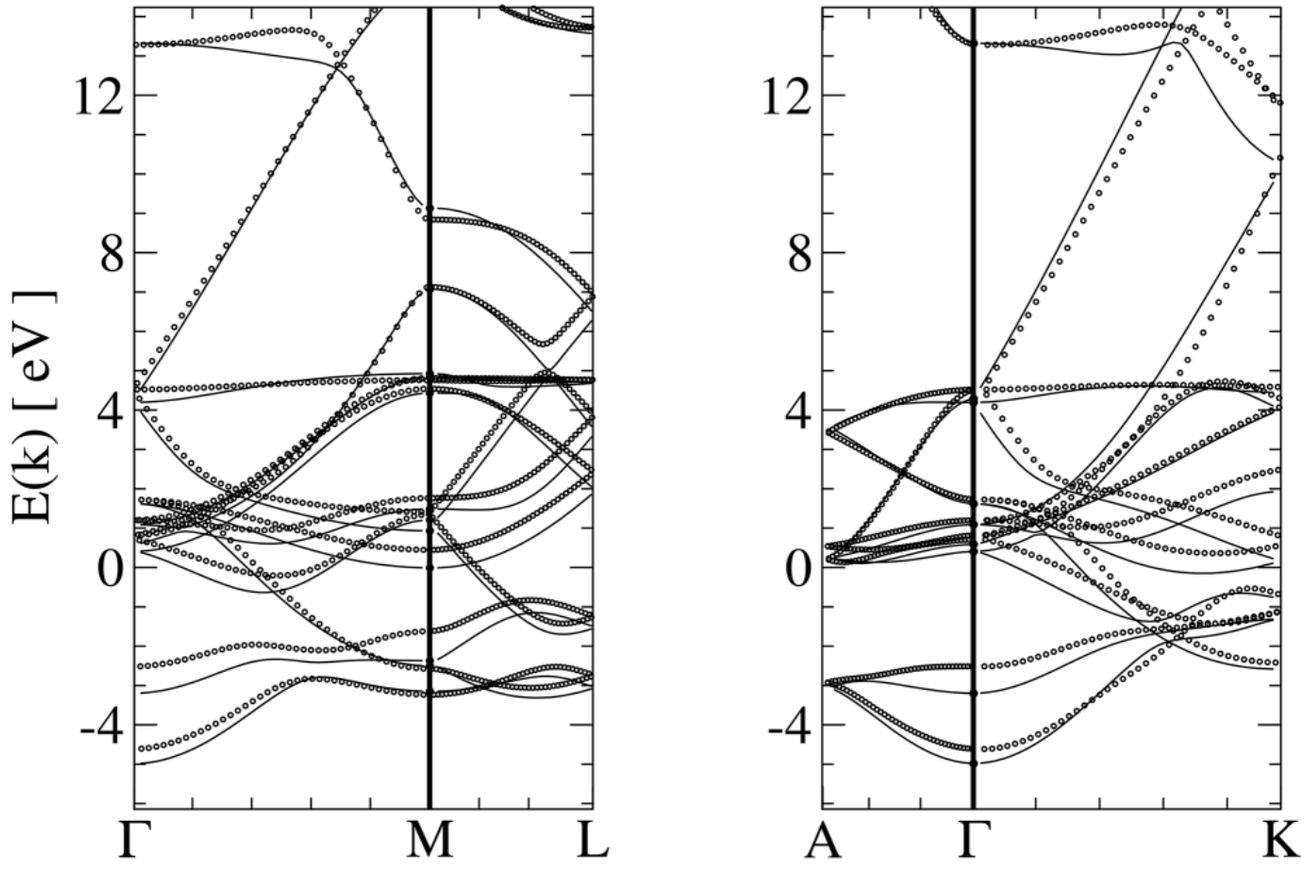
Hopping energy

- 9 orbitals per Ru site (4d, 5s, 4p) — [Kr] $4d^7 5s^1$

- Model parameters fit to DFT calculations obtained from Quantum Espresso

Electronic band structure fits

- Bloch states, TB and DFT band structures fit $|\psi_{\lambda\vec{k}}\rangle = \sum_{il,\kappa} c_{\lambda\vec{k},i\kappa} \exp\left(i\vec{k}\cdot\vec{R}_l\right)|\phi_{i\kappa}\rangle$
- Starting point were parameters for non-orthogonal TB model [5].
- Band structure fits matched symmetry representation of the bands [6].

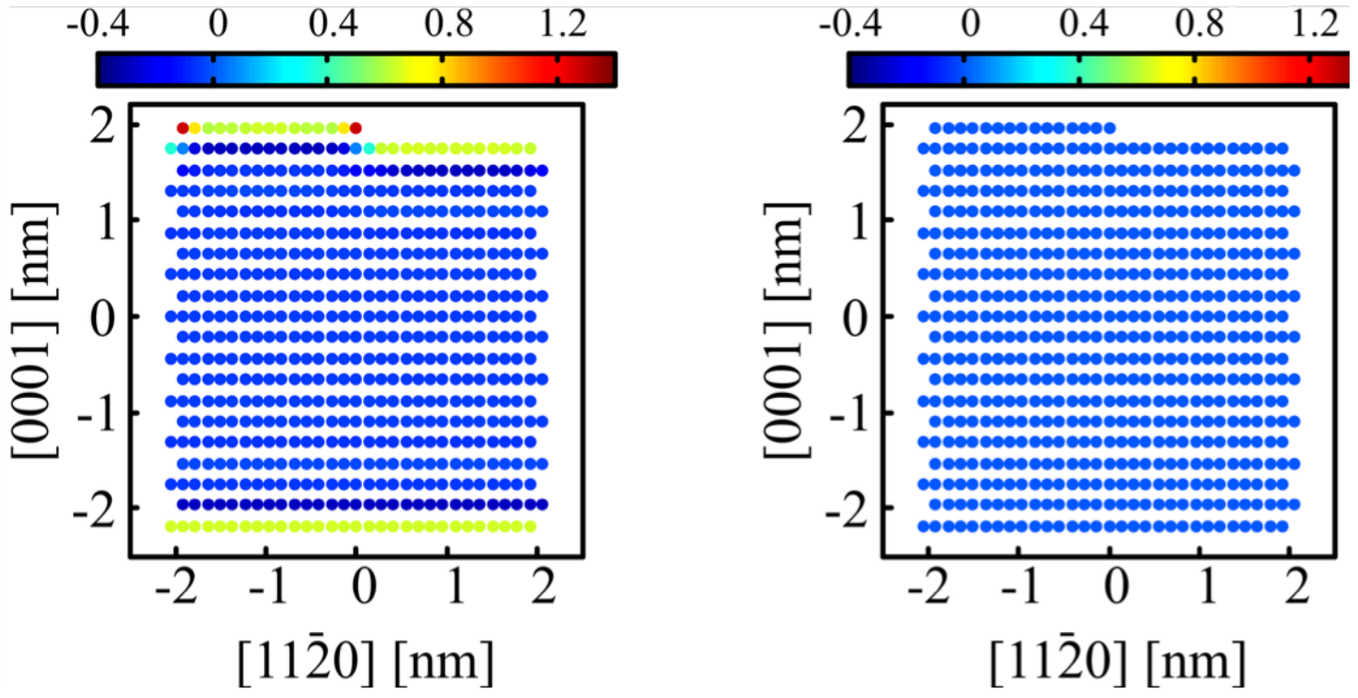


[5] M. J. Mehl *et al*, *Phys. Rev. B*.**54**, 4519 (1996)
[6] W. E. Richardson *et al*, ***J. Appl. Phys.* 130,** 195108 (2021)

Application of model to describe thin films

- The model determined from the bulk calculations not immediately transferable to surfaces — direct diagonalization of hamiltonian demonstrates excess electrons on surfaces and edges

- Lagrange multipliers added to onsite terms to impose local site neutrality



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Conductivity calculation using tight-binding electronic states

- The conductivity is evaluated use the Kubo-Greenwood equation

$$\sigma_{\alpha\beta}(E) = \frac{2\pi\hbar e^2}{\Omega}\mathbf{Tr}\left[\hat{v}_\alpha \delta(E-\hat{H})\,\hat{v}_\beta \delta(E-\hat{H})\right]$$

$$\hat{v}_\alpha = \frac{d\hat{x}_\alpha}{dt} = \frac{i}{\hbar}\left[\hat{H}, \hat{x}_\alpha\right]$$

- We use a basis of random initial vectors to evaluate trace,

$$\sigma_{\alpha\beta}(E) \approx \frac{2\pi\hbar e^2}{\Omega}\frac{1}{R}\sum_{i=0}^{R-1}\langle R_i|\,\hat{v}_\alpha\delta(E-\hat{H})\hat{v}_\beta\delta(E-\hat{H})\,|R_i\rangle$$

- Very efficient! We can handle millions of sites — 10's of nm scales

Kernel polynomial method — expansion of Dirac Delta functions into a Chebyshev polynomial series

$$|\Psi_{\alpha,i}(E)\rangle = \delta(E-\hat{H})\hat{v}_\alpha |R_i\rangle$$

$$|\Phi_{\beta,i}(E)\rangle = \hat{v}_\beta \delta(E-\hat{H}) |R_i\rangle$$

$$\sigma_{\alpha\beta}(E) \approx \frac{2\pi\hbar e^2}{\Omega}\frac{1}{R}\sum_{i=0}^{R-1}\langle\Psi_{\alpha,i}(E)|\Phi_{\beta,i}(E)\rangle$$

- We use a polynomial expansion of the Dirac delta functions — scales linearly with system size

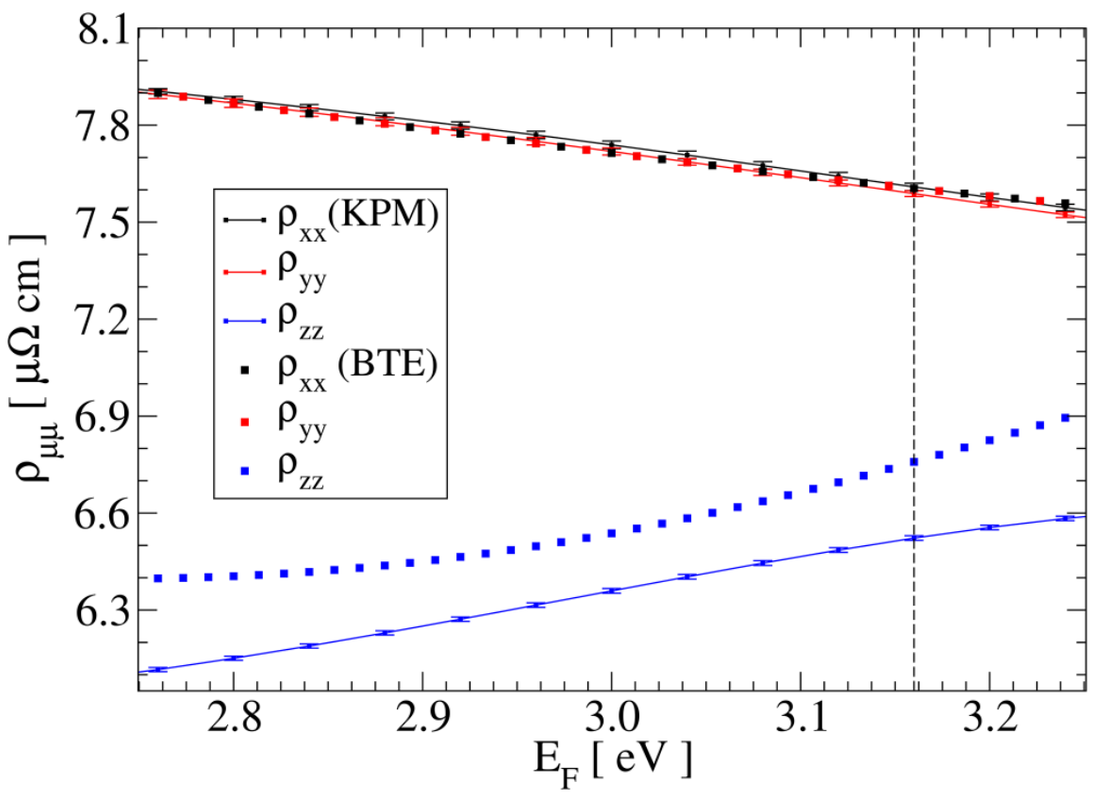
$$|\Psi_{\alpha,i}(E)\rangle = \frac{1}{\pi\sqrt{1-\varepsilon^2}}\left[g_0 + \sum_{n=1}^{N_T-1} g_n T_n(\varepsilon)\, T_n(\hat{h})\, \hat{v}_\alpha\right]|R_i\rangle$$

$$|\Phi_{\beta,i}(E)\rangle \frac{1}{\pi\sqrt{1-\varepsilon^2}}\left[g_0 + \sum_{n=1}^{N_T-1} g_n T_n(\varepsilon)\, \hat{v}_\beta\, T_n(\hat{h})\right]|R_i\rangle$$

Bulk transport, KPM vs. BTE using the tight-binding bands

- Good agreement between KPM calculations and BTE which depends only on band structure

- Results fit for scattering to obtain agreement with experiment — number of moments in KPM expansion scales linearly with computed resistivity



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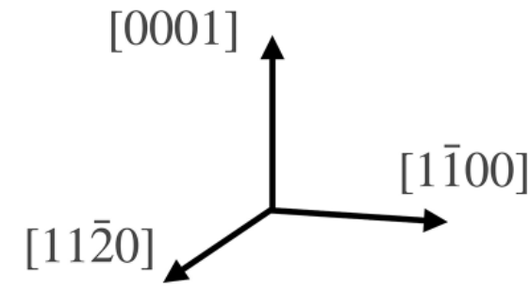
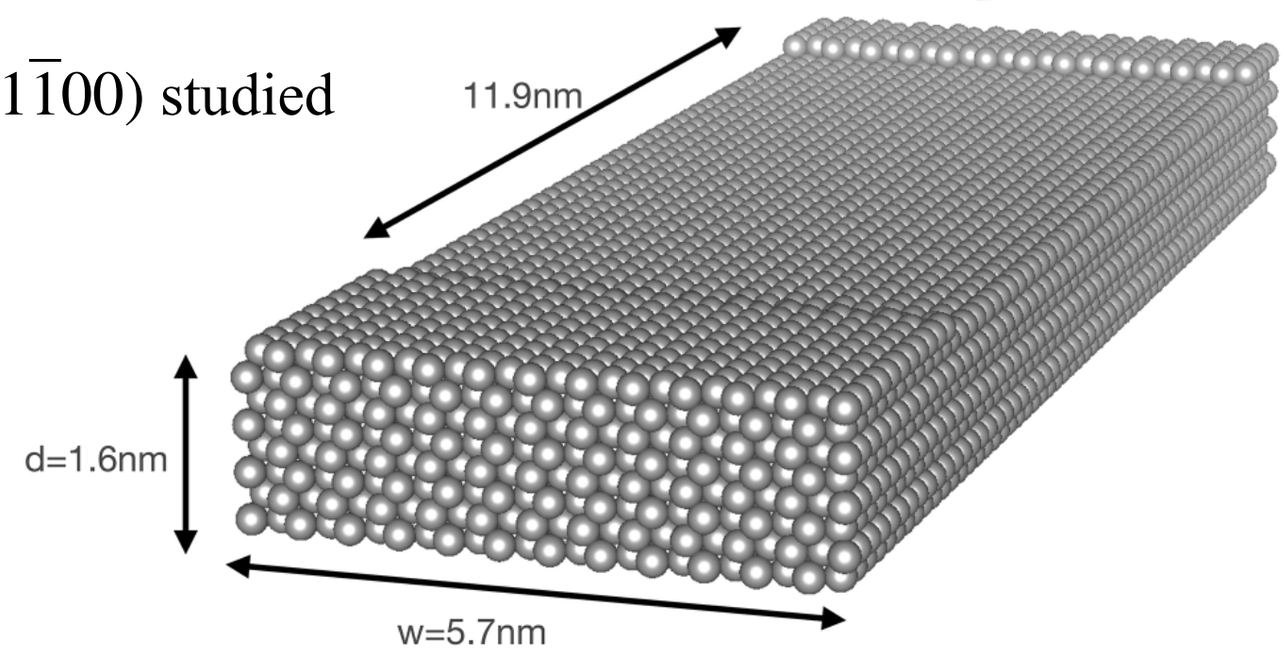
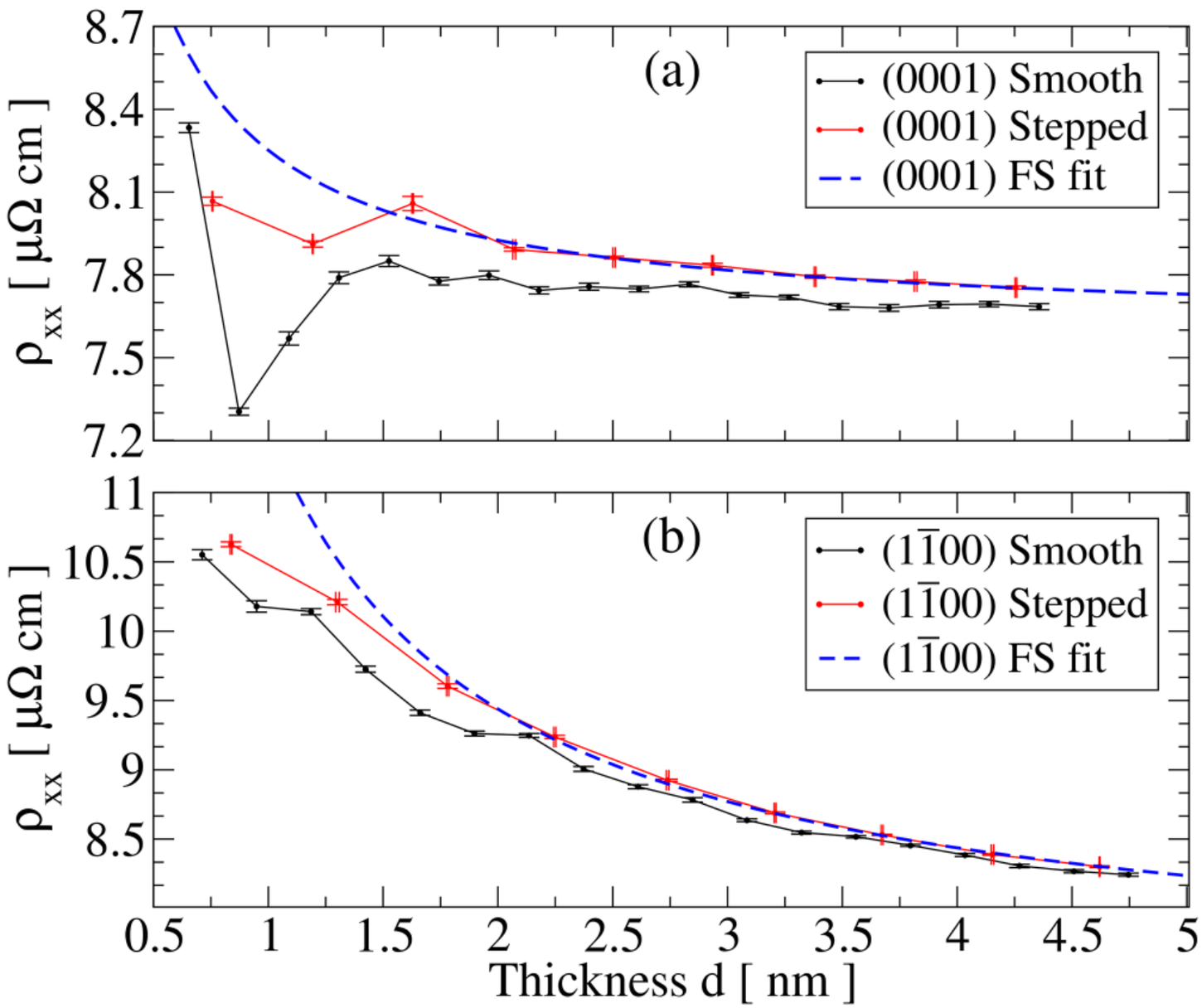
Thin film structure — steps ~ 10nm apart

- Film structures correspond closely to experiment
- Film ~ 140 nm long in the transport direction
- Transport direction [11$\bar{2}$0]
- Two film orientations, (0001) and (1$\bar{1}$00) studied





Results — two different surface orientations



Theoretical interpretation of results

- We followed the analysis previously published for films studied using non-equilibrium Green's functions (NEGF) [7].

- Resistivity is determined from a "flat film" term with added terms due to scattering from steps with transmission probability $\eta(s)$ which depends on step height s

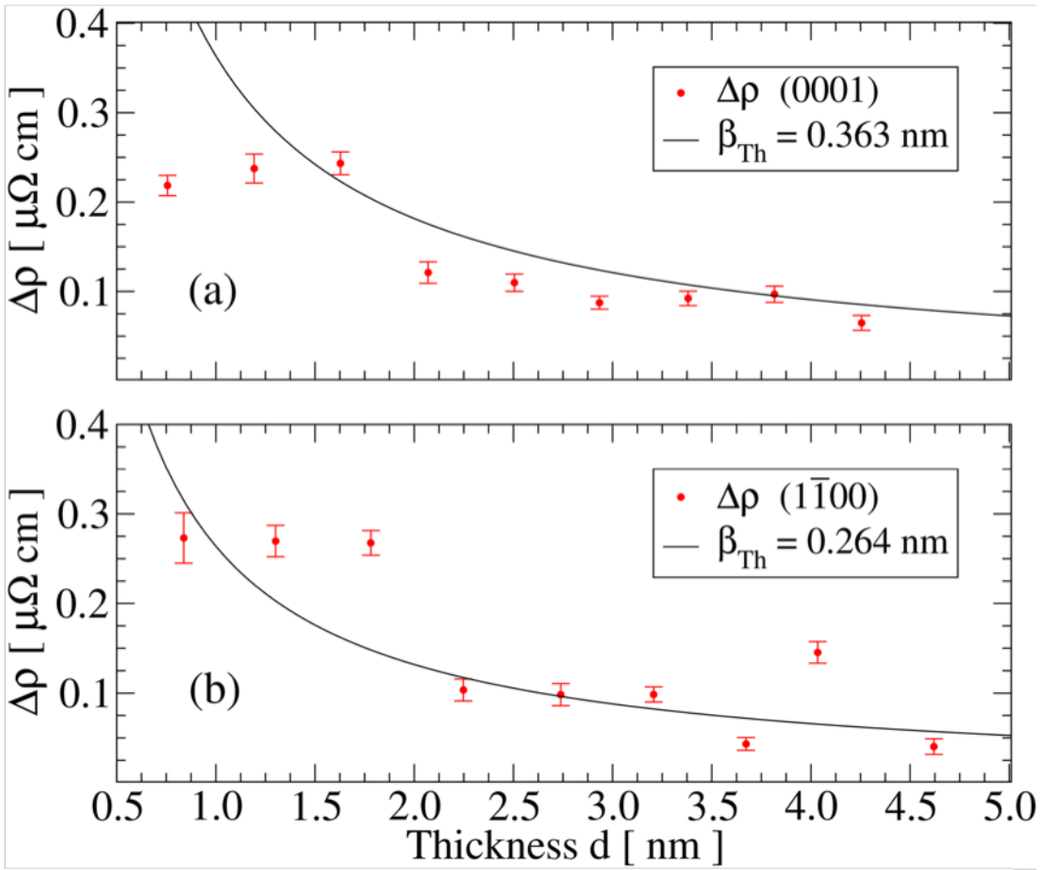
$$\rho = \rho_{ff} + \frac{1}{g_0 L}\sum_{i=1}^{N}\left(\frac{1}{\eta_i} - 1\right)$$

- NEGF found dependence on step height,

$$\eta(s) = 1 - \frac{s}{d}$$

- For our model we compute resistivity change,

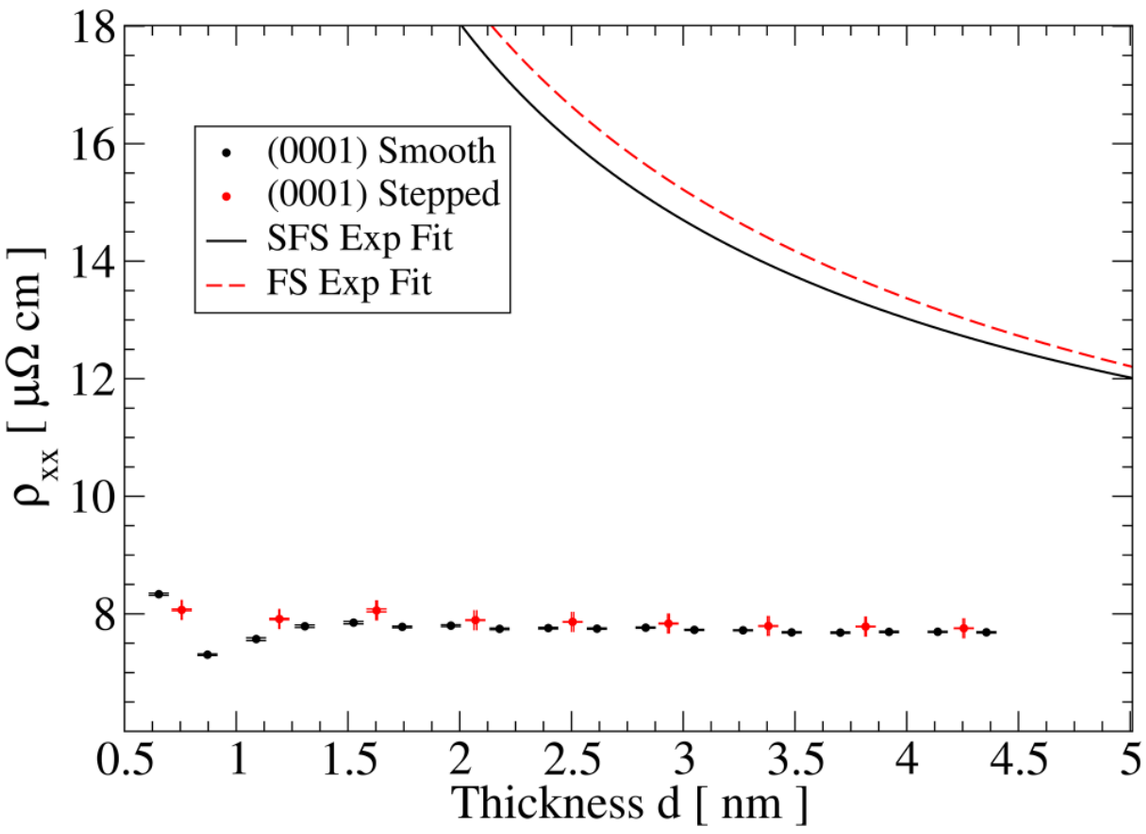
$$\Delta\rho = \frac{\beta_{Th}}{d}$$

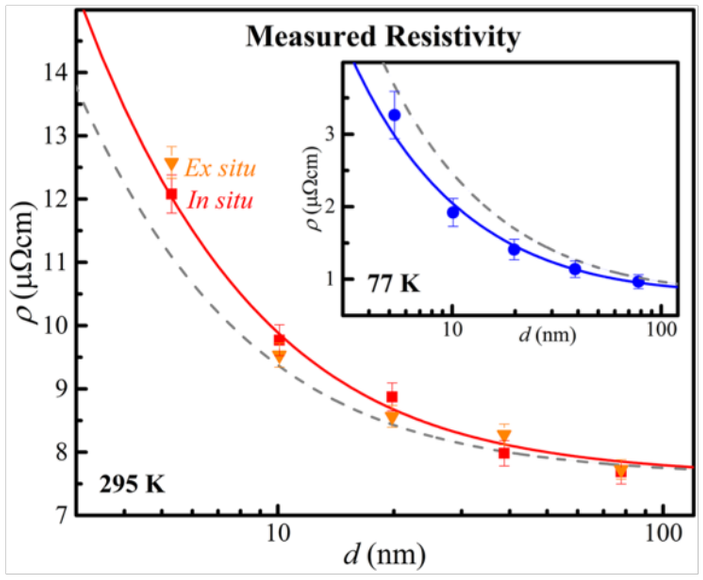


[7] T. Zhou *et al*, *J. Appl. Phys.***123**, 155107 (2018)

Comparison to experiment

- Very strong surface scattering not reproduced by surface steps
- Contribution due to stepped surfaces very minimal

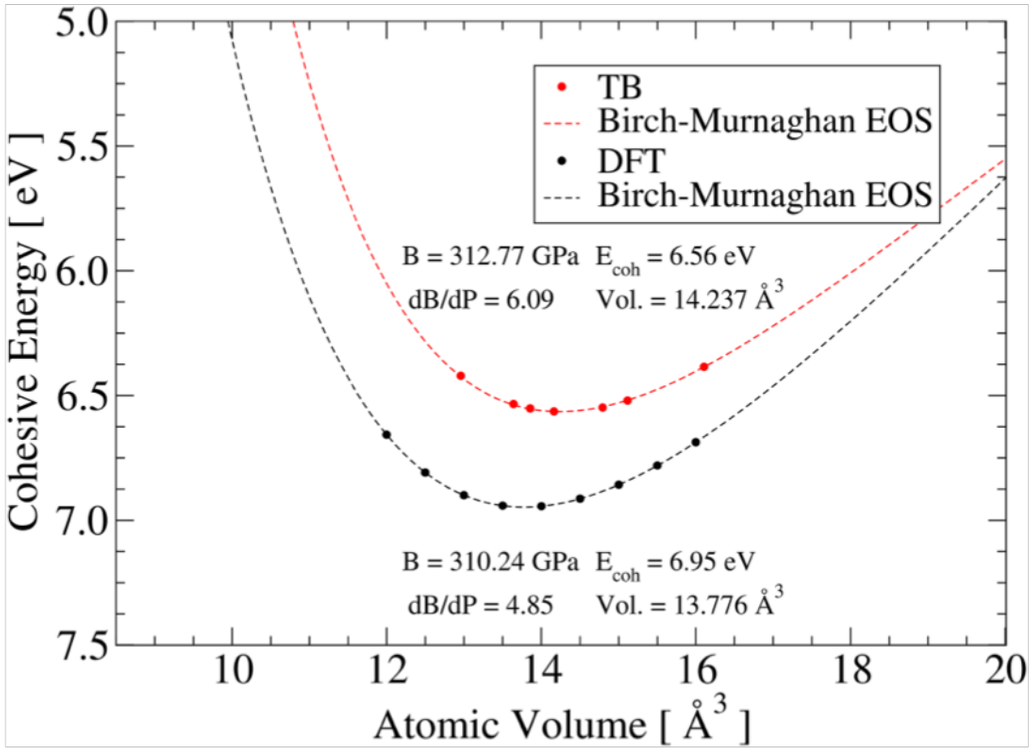




Milosevic et al, *J. Appl. Phys.* **124**, 165105 (2018)

Size-dependent electron-phonon scattering?

- Currently working on phonons, electron-phonon scattering
- Model also determines cohesive energy, agrees with DFT results
- Surface atoms less strongly bonded (fewer neighbors) likely have lower frequency vibrational states, larger amplitude, perhaps scatter more effectively



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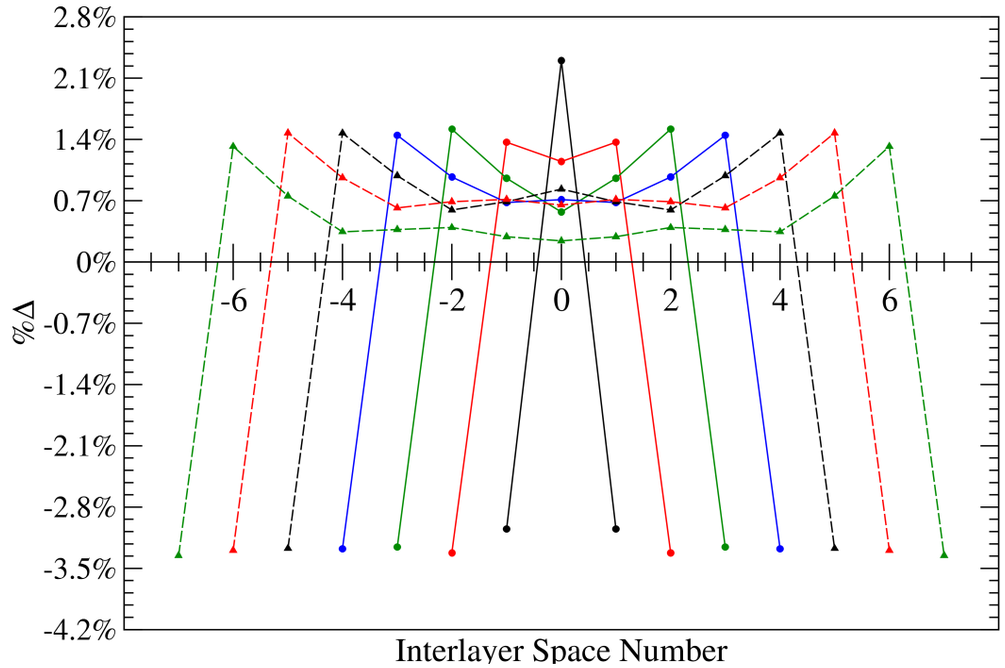
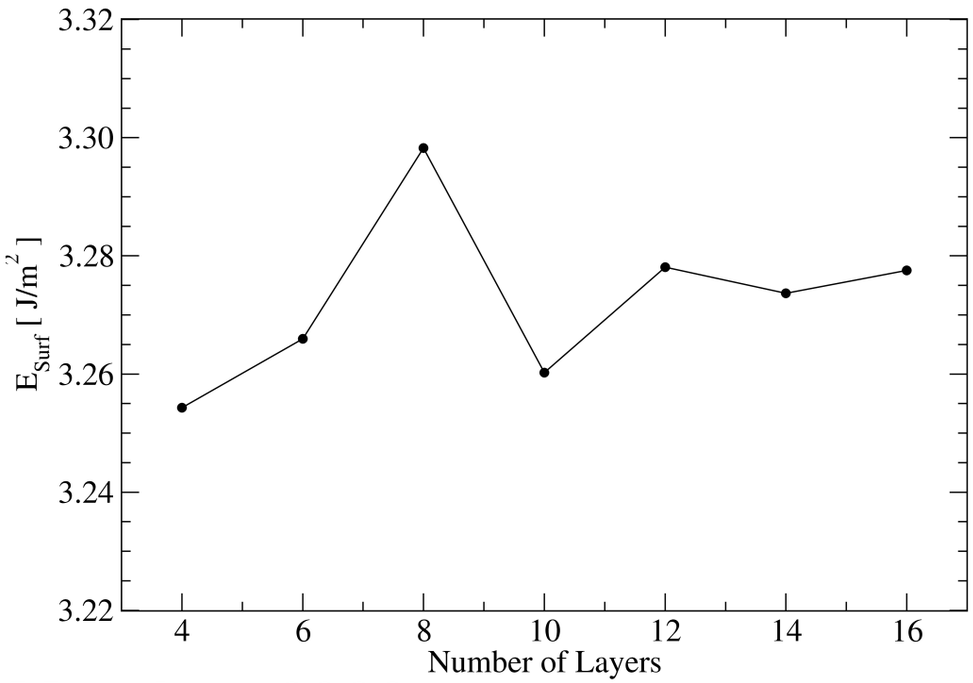
Surface relaxation, (0001) Ru

- Inward relaxation of outer layer, comparable to ~3% relaxation in DFT.
- Converged surface energy, comparable to range of values 2.6-2.9 Jm^{-2} from DFT.
- Surface phonons, phonon band structures being computed
- Transport computed as ensemble average over Bose-Einstein occupation of phonon states.





Conclusions

- Large-scale transport calculations possible with realistic TB models fit to DFT — KPM method for transport.

- Application to scattering at steps in Ru thin films — steps only make a minor contribution

- Electron-phonon scattering currently under investigation — might yield thickness-dependent scattering

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