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DC Conductivity of Platinum From Ab Initio Simulation

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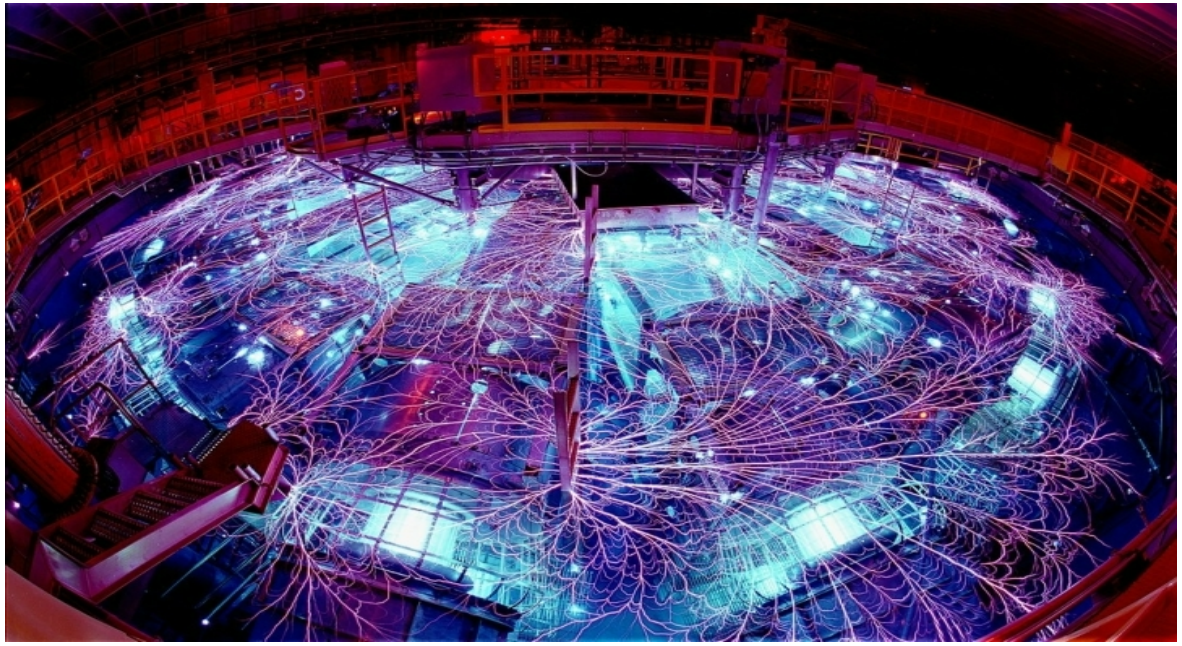
Motivations

Platinum used as high pressure standard for shock compression experiments on the Z machine

These experiments subjected to very large currents and magnetic fields



The conductivity of the standard needs to be known

Experiments are difficult in thermodynamic regimes relevant for Z - ab inito theory is crucial

Molecular Dynamics

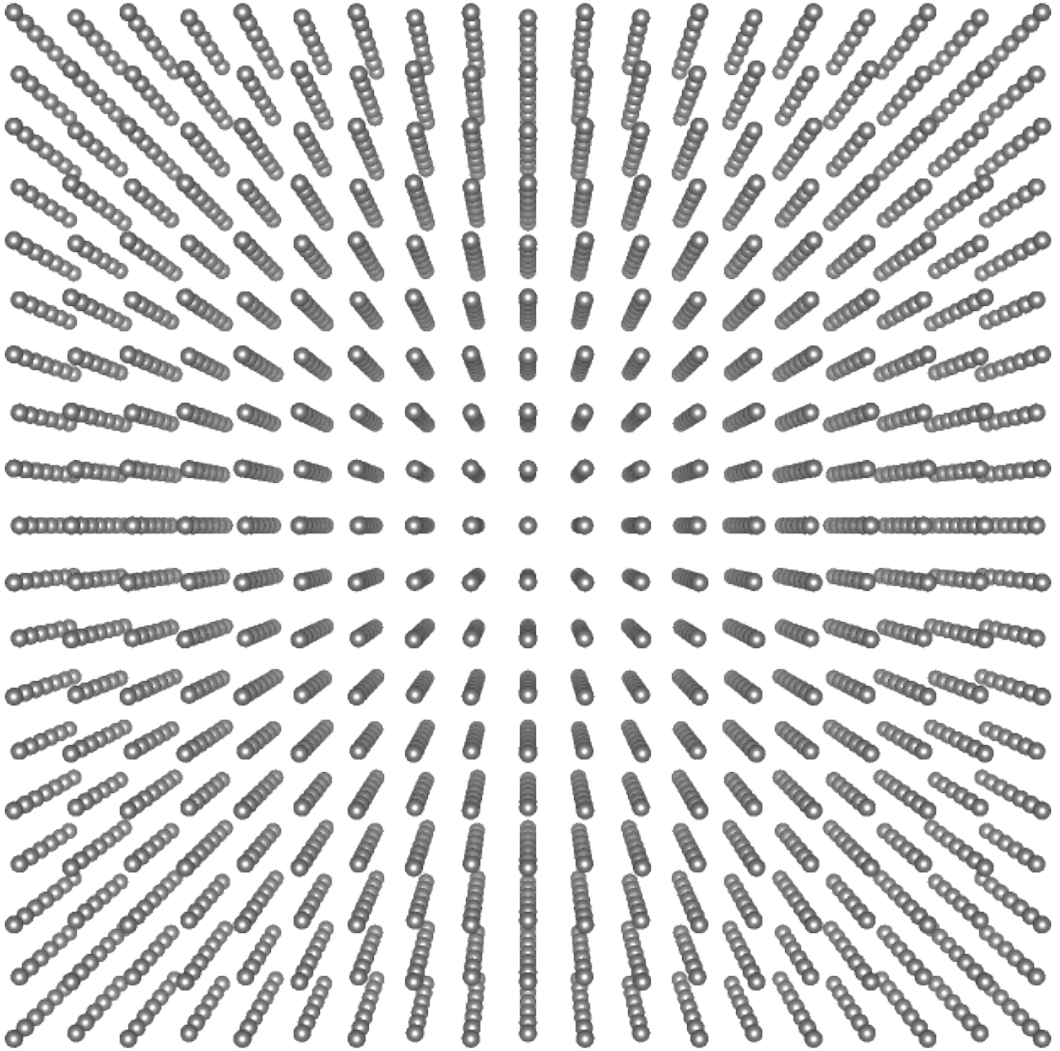
Vienna Ab Initio Simulation Package

- DFT – PBE
- PAWs
- 10 e^- Pseudopotential

  [Xe] $4f^{14}\ 5d^{9}\ 6s^{1}$
- NVT ensemble, velocity-scaling thermostat
- Sampled at Γ point

108 atom supercell

15,000 $1fs$ timesteps



Density Functional Theory

- The external potential (and total energy) are unique functionals of the electron density.
- The density that minimizes the total energy is the exact ground state density.

$$E[\rho(r)] = F_{XC}[\rho(r)] + \int \rho(r)\ V_{ext}(r)dr$$

- $F_{XC}[\rho(r)]$ unknown universal functional, must be approximated

$$F_{XC}[\rho(r)] = T_s[\rho(r)] + E_H[\rho(r)] + E_{XC}[\rho(r)]$$

- Solving N single-particle differential equations

Electrical Conductivity – Kubo Greenwood (KG)

$$\sigma_k(\omega) = \frac{2\pi e^2 \hbar^2}{3m^2 \omega \Omega} \sum_{j=1}^{N} \sum_{i=1}^{N} \sum_{\alpha=1}^{3} \left[F(\epsilon_{i,k}) - F(\epsilon_{j,k}) \right] \left| \langle \Psi_{j,k} | \nabla_\alpha | \Psi_{i,k} \rangle \right|^2 \delta(\epsilon_{j,k} - \epsilon_{i,k} - \hbar\omega)$$

N discrete bands, Ω cubic supercell volume element, F Fermi weight, Ψ electronic wave function.

VASP KG simulations – average over many snap shots, x-y-z-components

M. P. Desjarlais, J. D. Kress, and L. A. Collins Phys. Rev. E **66**, 025401(R) 26 August 2002.

DC Conductivity

Ambient conditions, $\rho = 21.45\ \frac{g}{cm^2}$

Sampled at 4x4x4 Monkhorst

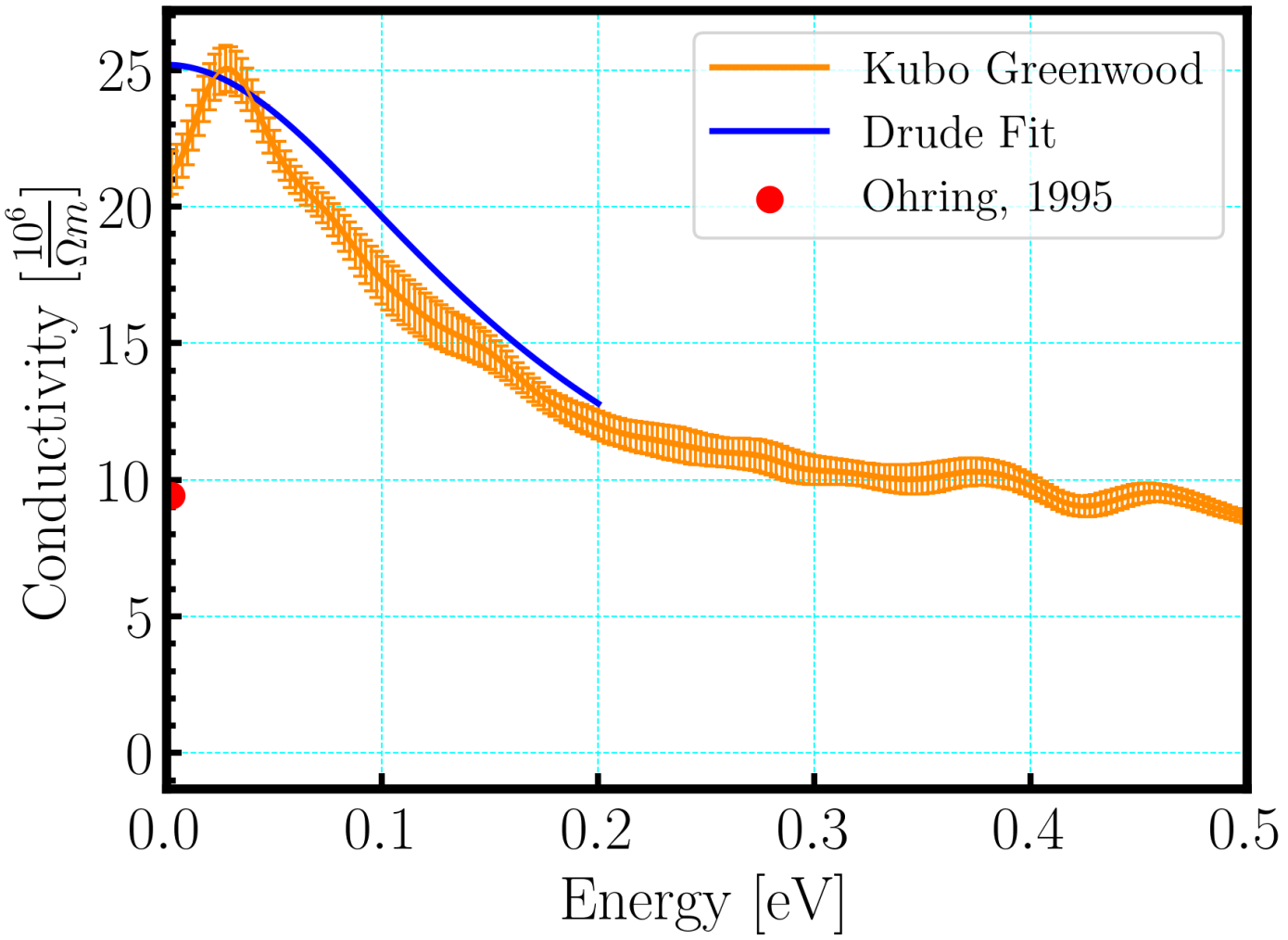
Irreducible Wedge

Four Snapshots

3,000 Discrete Bands

Fit low energy to Drude Model

$$\sigma(\omega) = \frac{\sigma_0}{1 + \omega^2\tau^2} + constant$$



Ohring, Milton. "Engineering Materials Science." New York: Academic Press, 1995.

Fermi Surface

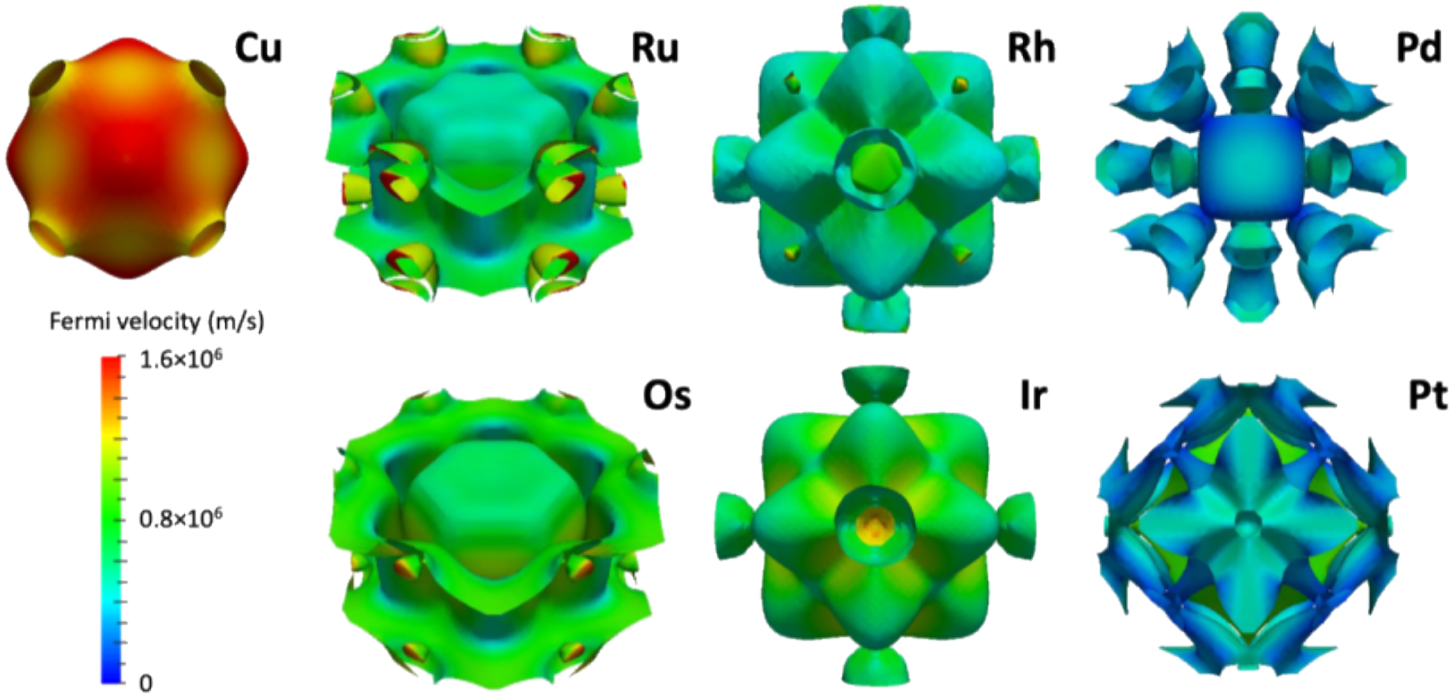


FIG. 4: Fermi surfaces of platinum-group metals. The Fermi surface of Cu is also shown as a reference. The color scheme indicates the Fermi velocity.

k points used determine where on the Fermi surface contributes to the conductivity.

Simple crystalline systems (Cu) with highly symmetric fermi surface require few **k** points.

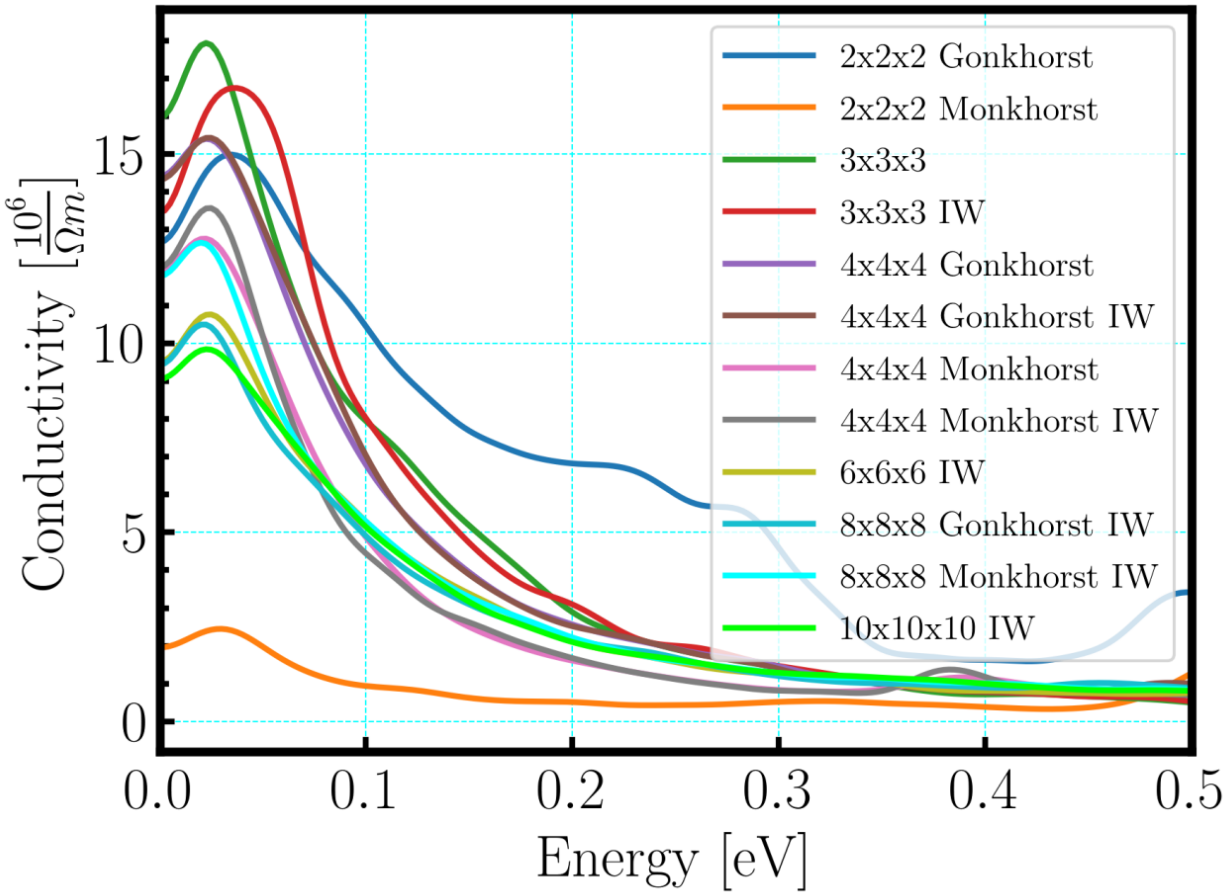
Platinum much more complex, need a complex **k** point mesh.

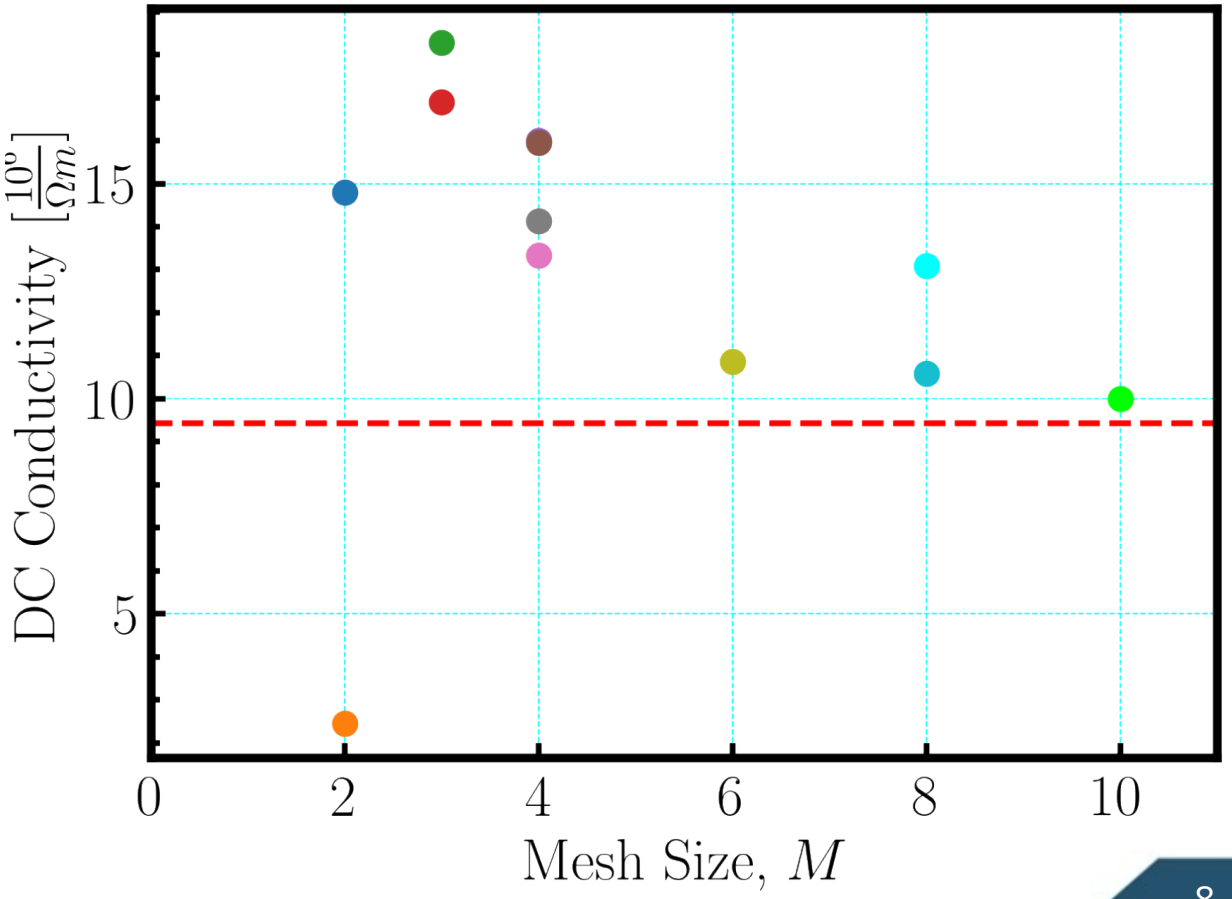
Dutta, Shibesh et al. "Thickness dependence of the resistivity of platinum-group metal thin films." *Journal of Applied Physics* 122 (2017): 025107.

Brilloiun Zone Sampling

Number of k points increase as M^3 for $M \times M \times M$ mesh

Irreducible wedge reduces computation costs.

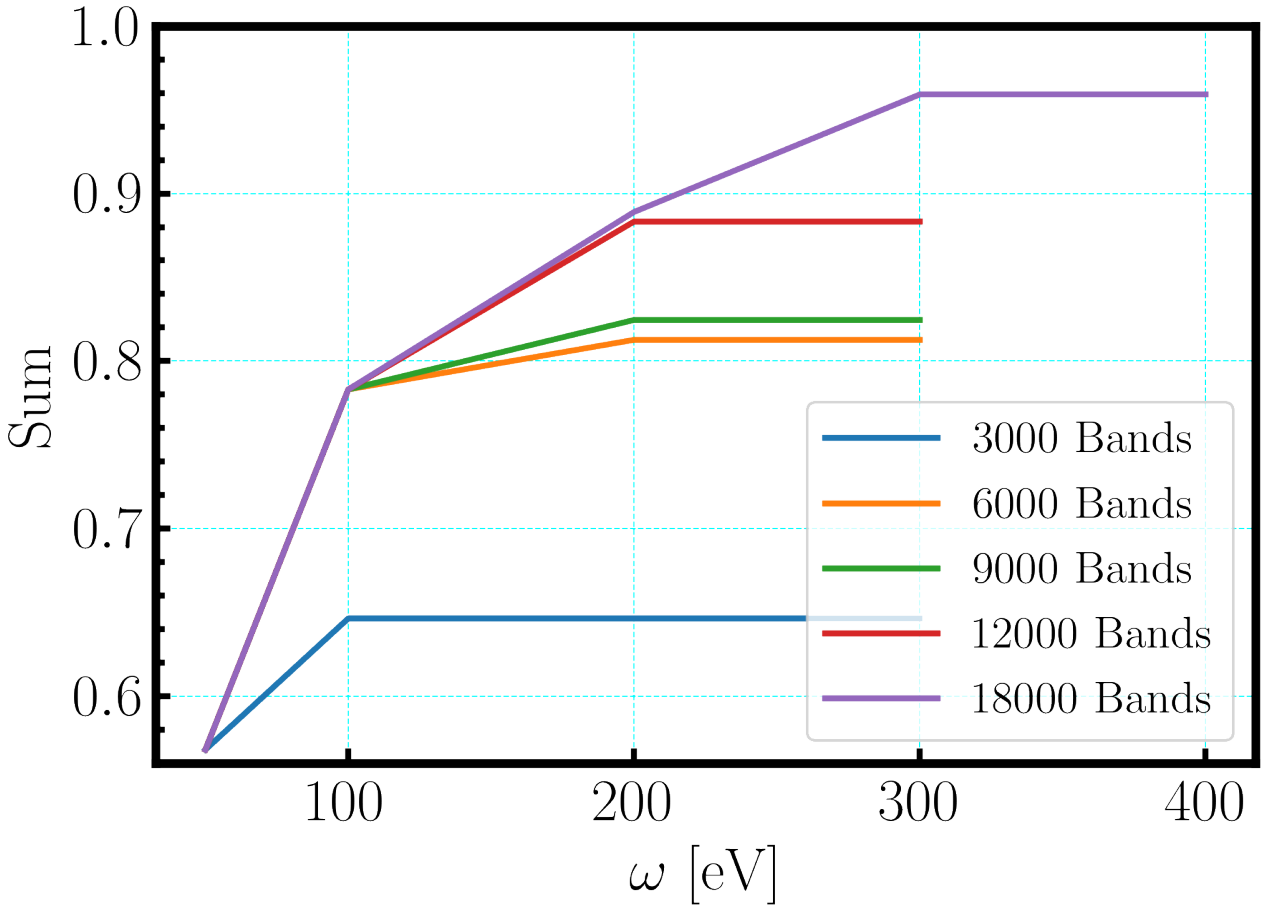


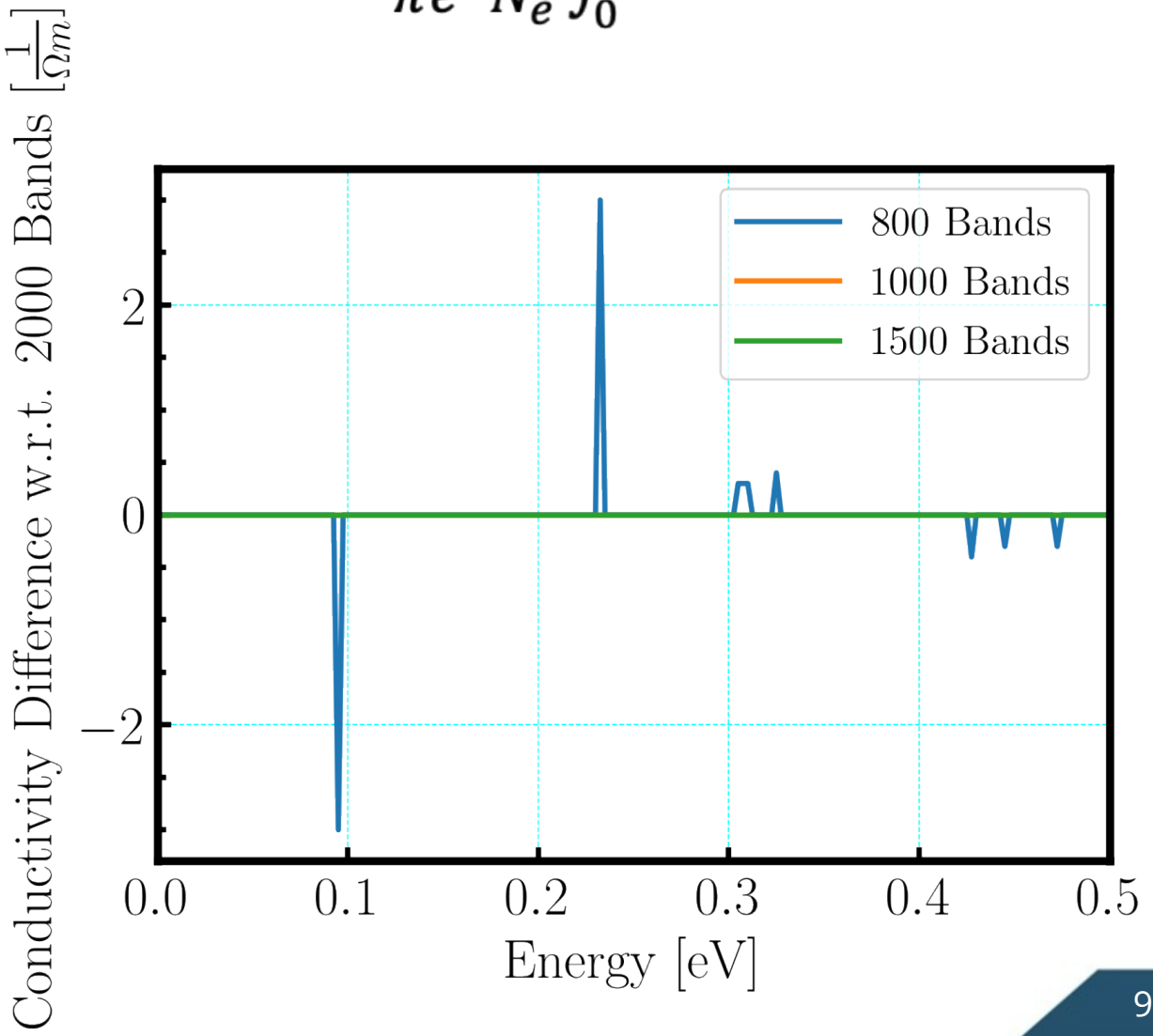


Number of Orbitals

DC conductivity converges much quicker than sum rule with increasing band number

$$S = \frac{2m\Omega}{\pi e^2 N_e}\int_0^\infty \sigma(\omega)d\omega = 1$$





Discrete Band Structure Smearing

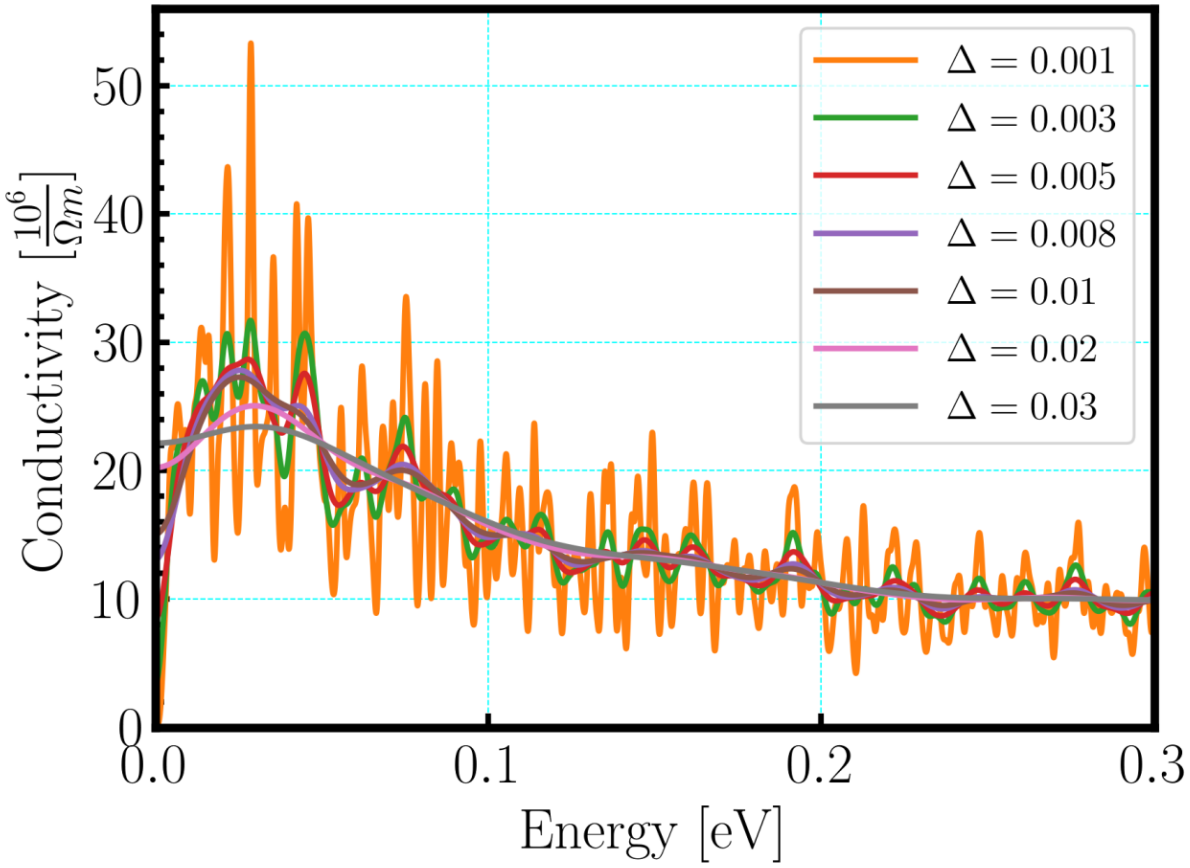
$$\sigma_k(\omega) = \frac{2\pi e^2 \hbar^2}{3m^2 \omega \Omega} \sum_{j=1}^{N} \sum_{i=1}^{N} \sum_{\alpha=1}^{3} \left[F(\epsilon_{i,k}) - F(\epsilon_{j,k}) \right] \left| \langle \Psi_{j,k} | \nabla_\alpha | \Psi_{i,k} \rangle \right|^2 \delta(\epsilon_{j,k} - \epsilon_{i,k} - \hbar\omega)$$

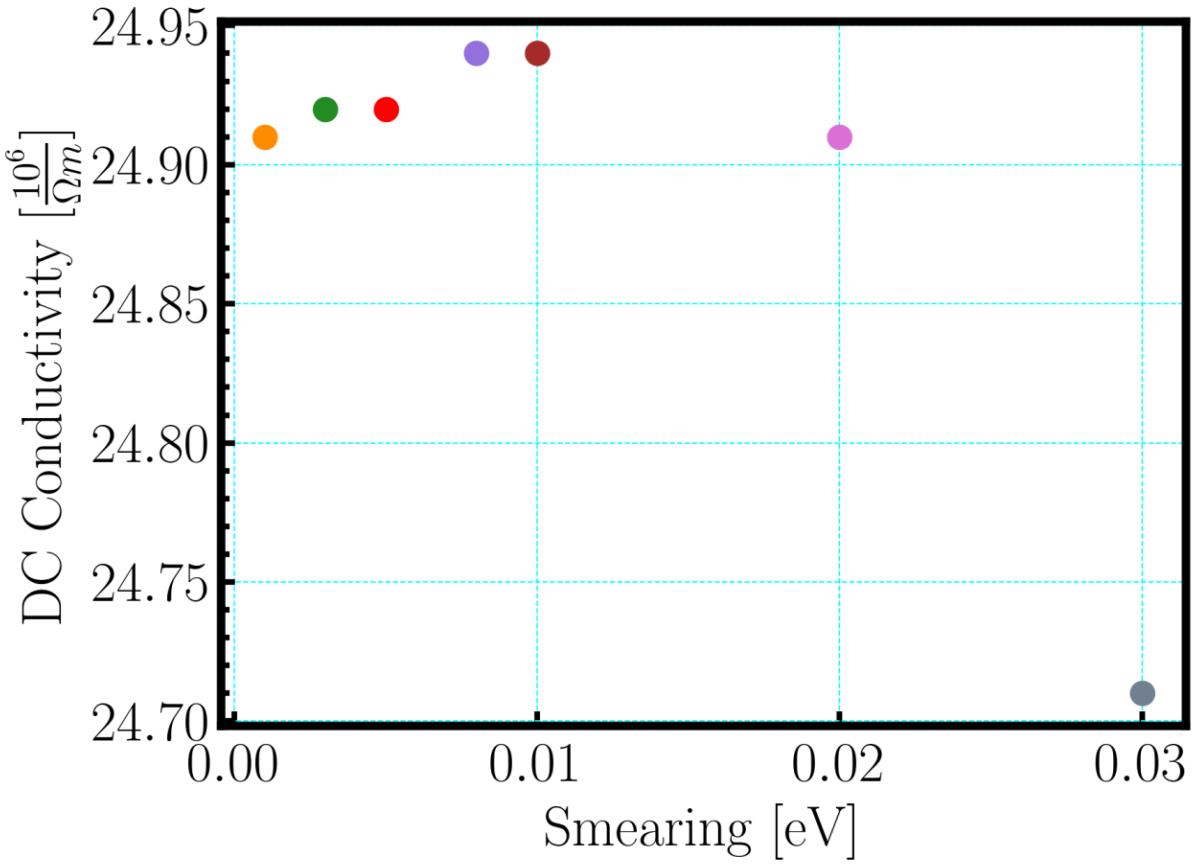
Apply Gaussian broadening – control width of Gaussian

Rule of thumb: start with energy difference between eigenvalues above and below Fermi energy

Discrete Band Structure Smearing Con't

Smooth out local oscillations without losing structure



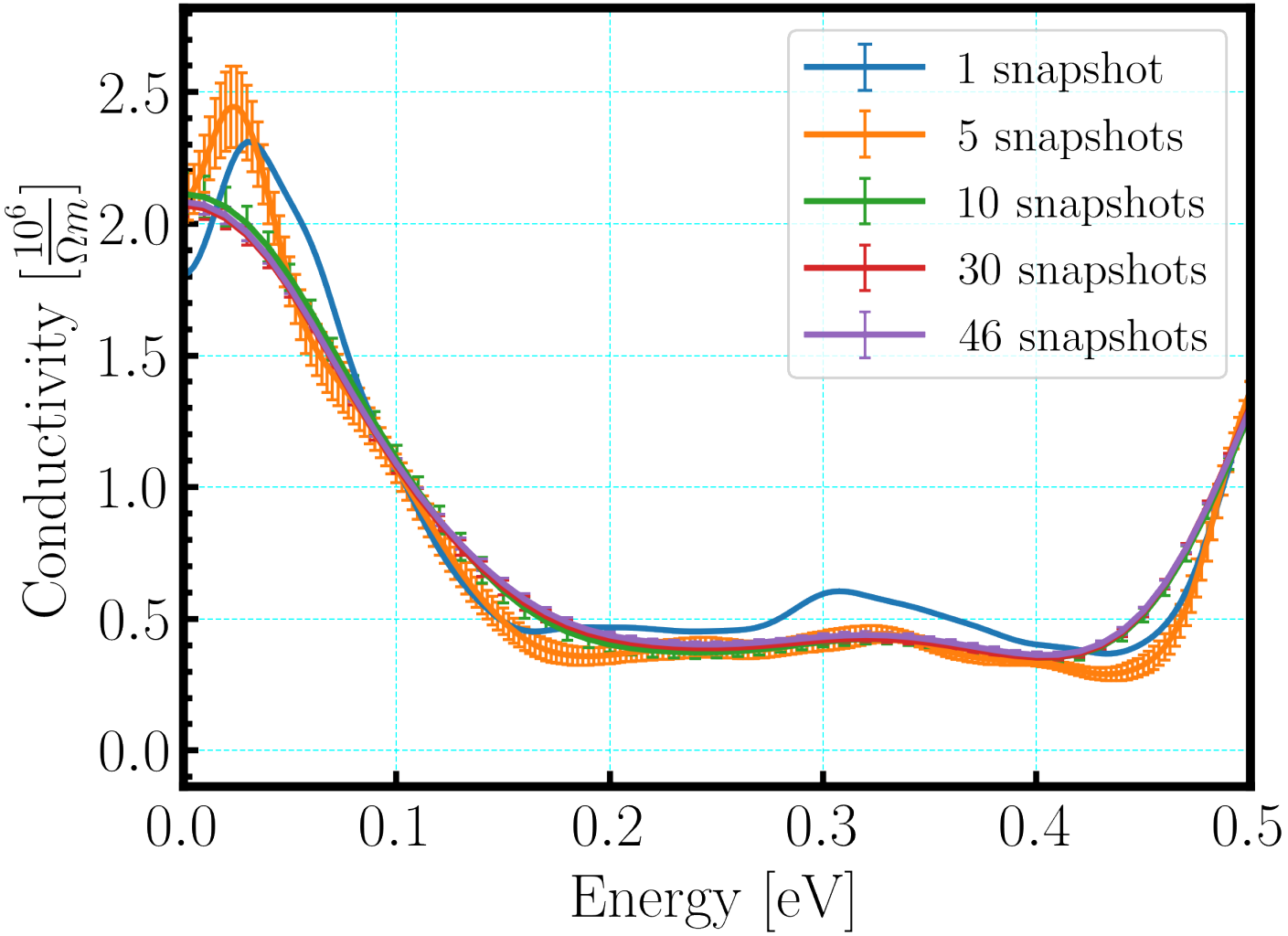


Sampled at 4x4x4 Monkhorst Irreducible Wedge

Number of Snapshots

Taken from end of MD simulation to ensure equilibrium

Snapshots spaced by nearest prime number to twice the correlation time



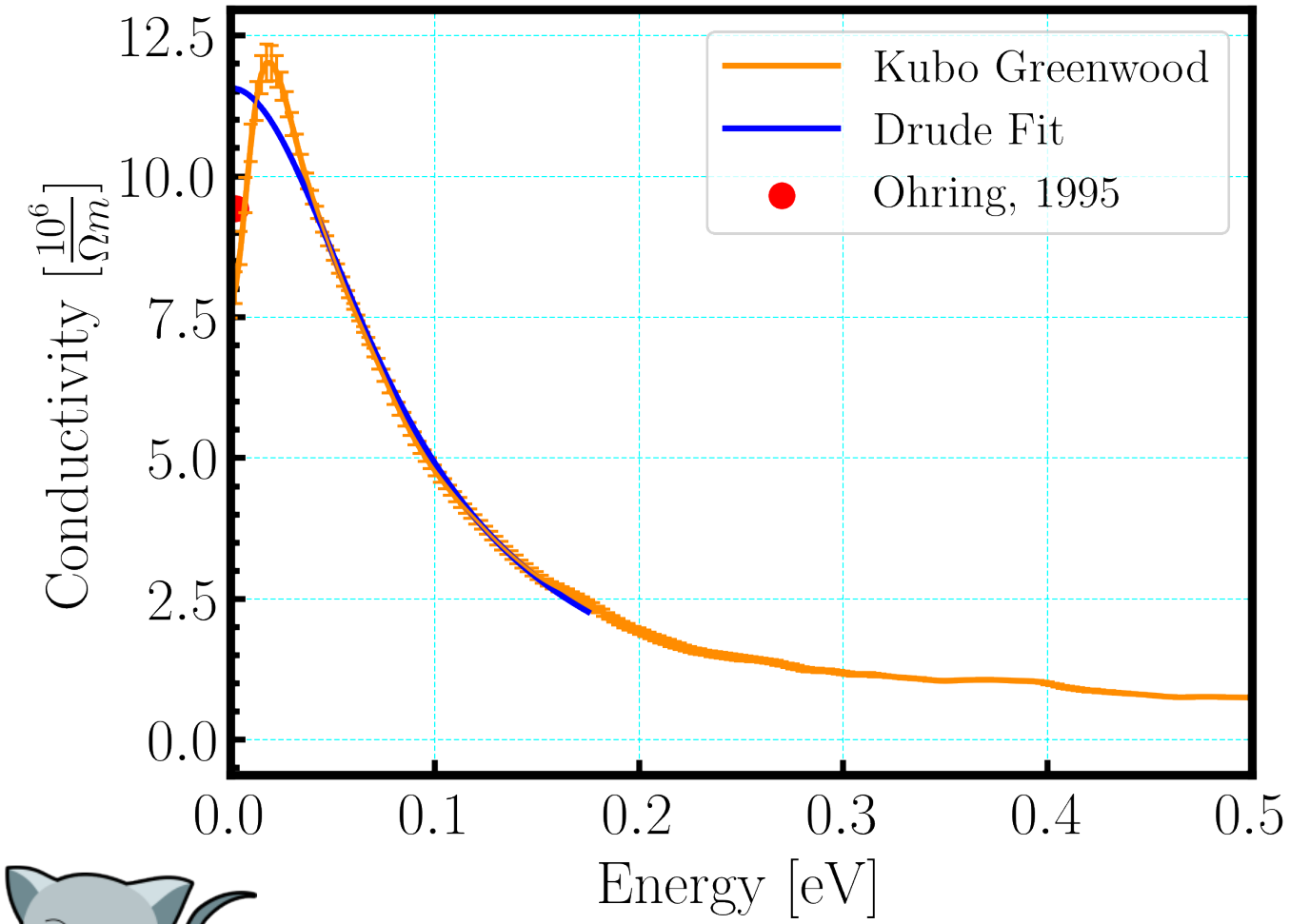
Conclusions

Implementing results from all convergence studies

$$\sigma_{DC} = 11.57 \frac{10^6}{\Omega m}$$

Within 23% of measured value,

$$\sigma_{0,exp} = 9.434 \frac{10^6}{\Omega m}$$



Future Work

- Extend to melt, vapordome
- Finite size effects will be quantified

Ohring, Milton. "Engineering Materials Science." New York: Academic Press, 1995.