LSQUANT: Linear Scaling Quantum Transport Methodologies

José H. Garcia

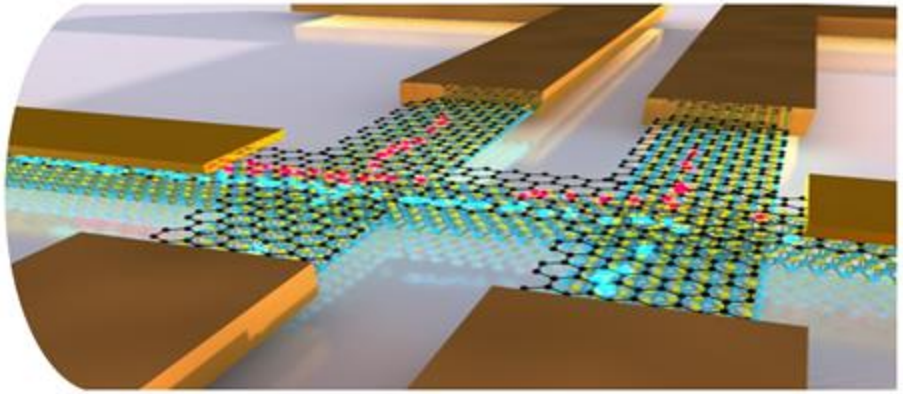
Barcelona – Jun 12th, 2022

Institut Català de Nanociència i Nanotecnologia

The goals of this presentation

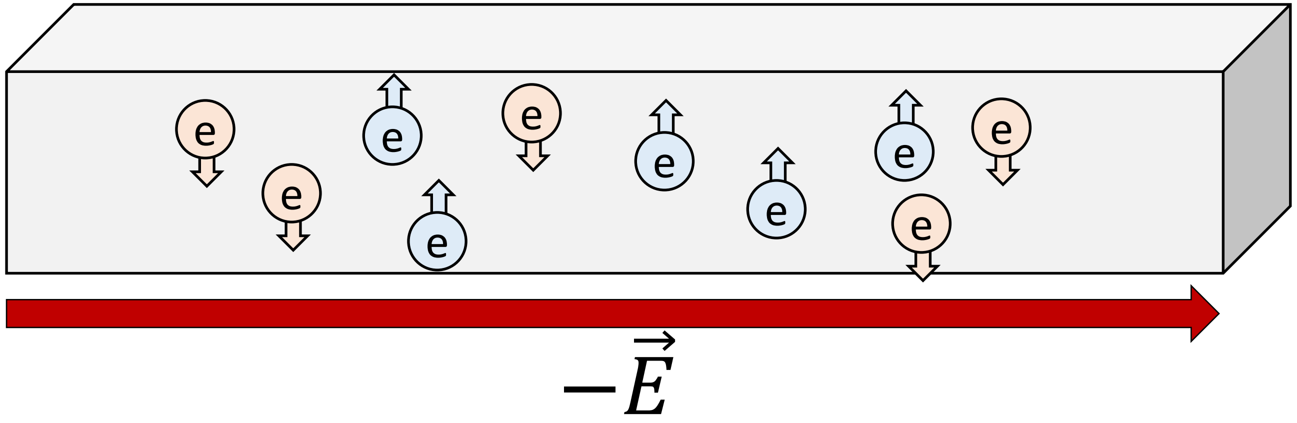
- Gain insights into the importance of electrical response in modern materials science.
- Understand the relationship between the Kubo Formula and electrical response.
- Learn how to numerically solve the Kubo formula in a linear-scaling algorithm.
- Present the up-and-coming LSQuanT package

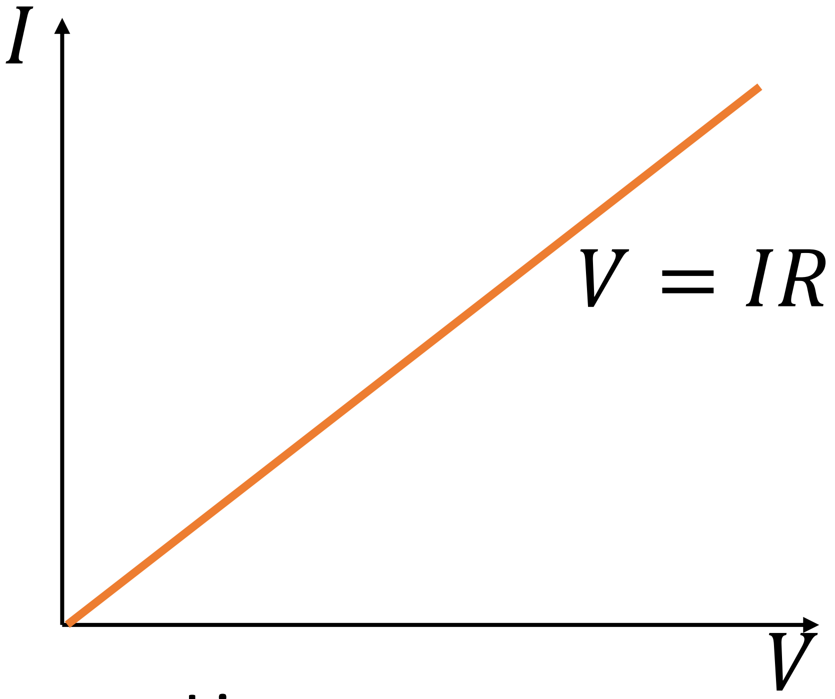
LSQUANT
LINEAR SCALING QUANTUM TRANSPORT



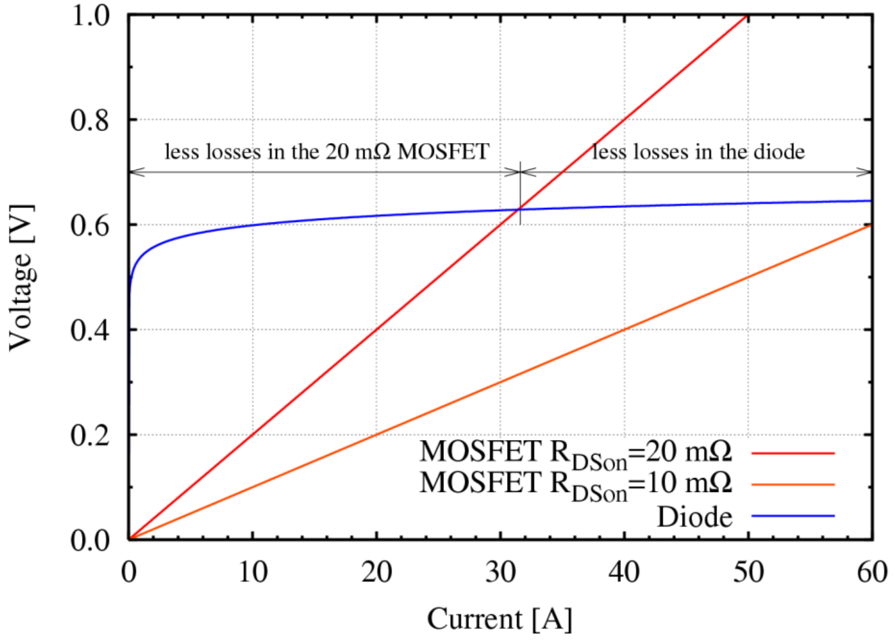
https://www.lsquant.org/lsq/index.php

Electrical Response



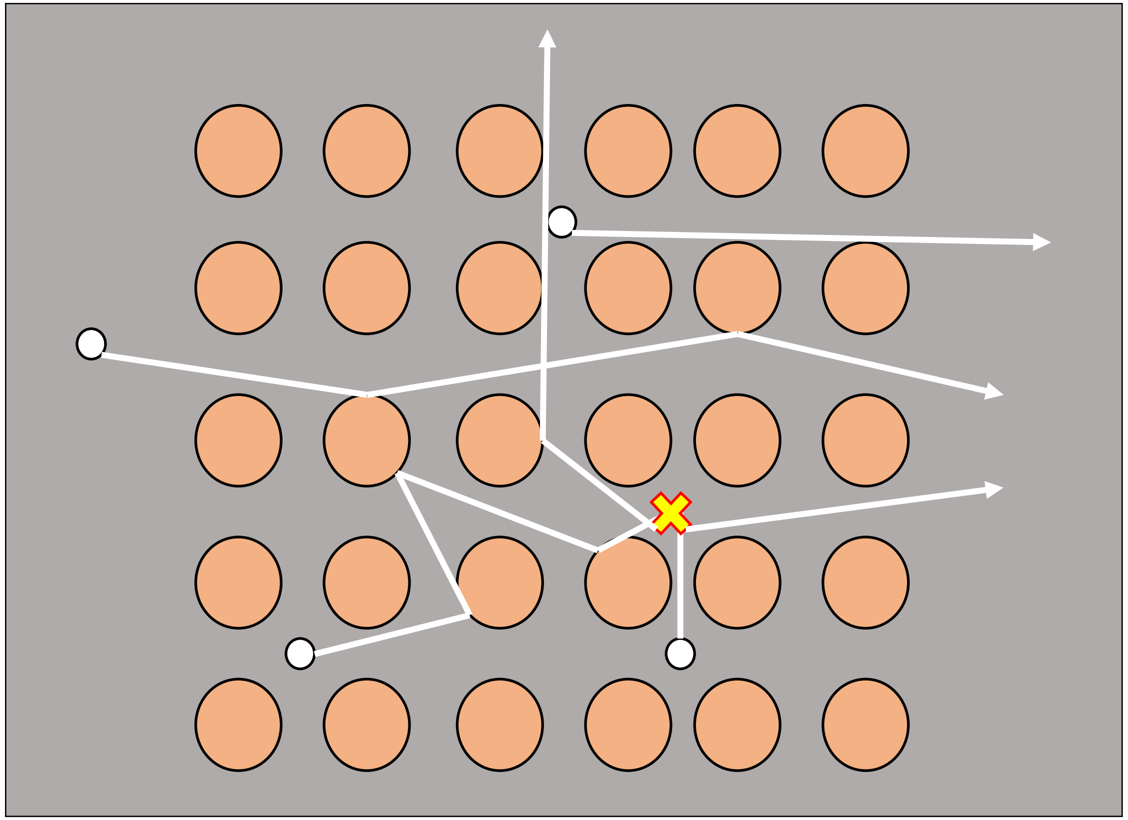


Linear-response



Nonlinear devices

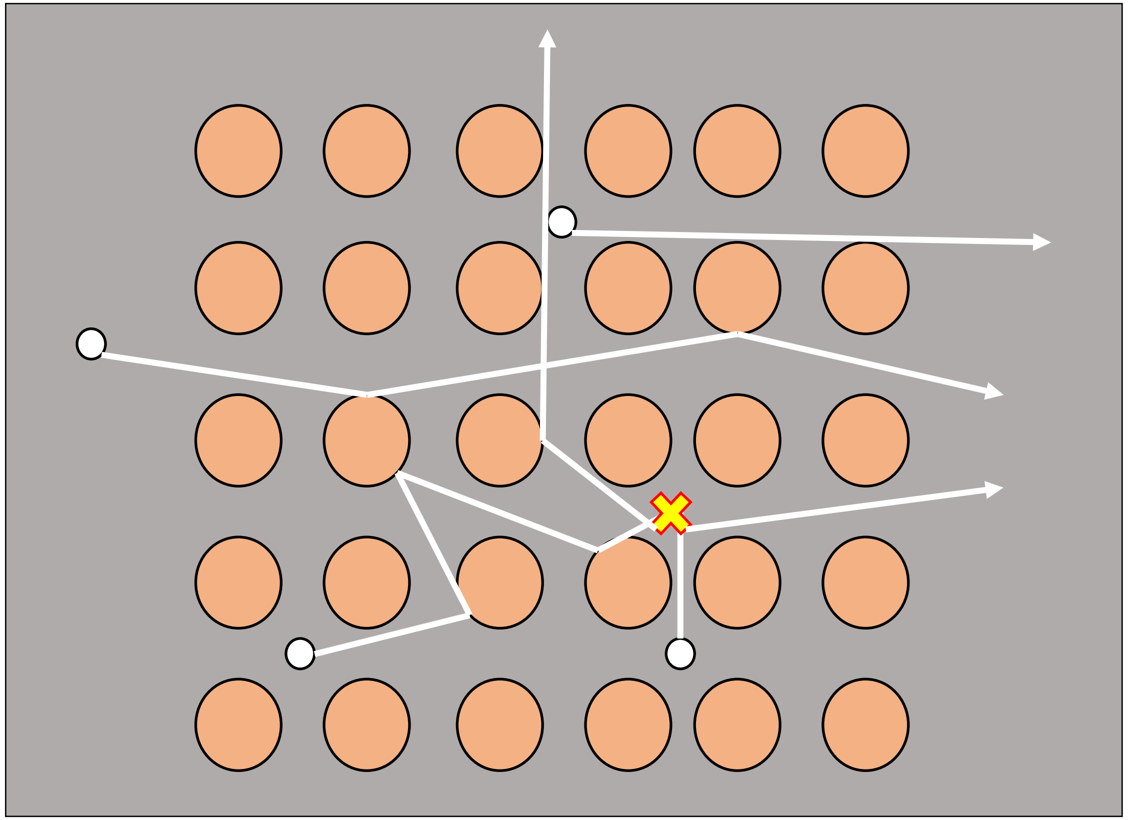
Dissipation



$$\frac{d\vec{p}}{dt} = q\vec{E}$$

Unlimited momentum increment

Dissipation



$$\frac{d\vec{p}}{dt} = \mathrm{q}\vec{E} - \frac{\vec{p}}{\tau}$$

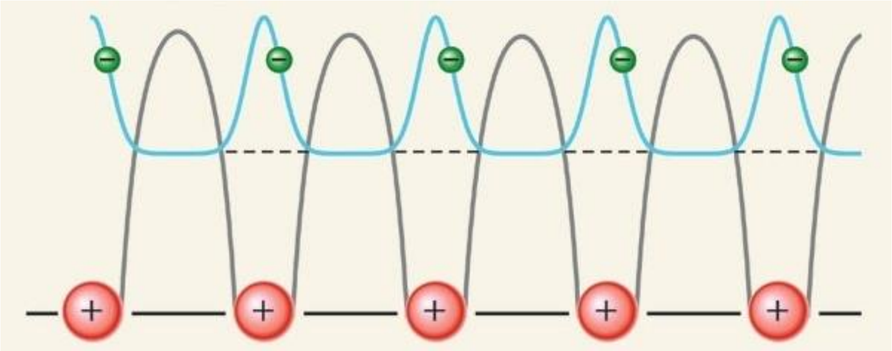
~~**Unlimited momentum increment**~~

$\boldsymbol{\tau}$

momentum relaxation time

$$\vec{J} = \frac{nq^2\tau}{m}\vec{E} \text{ (Drude model)}$$

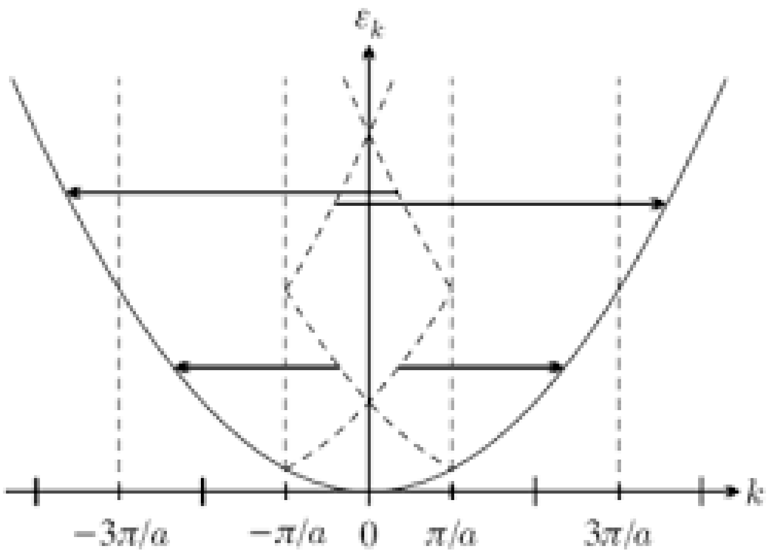
Quantum mechanics



Periodic potential

Bloch's theorem

Nearly free electros



$$\vec{p} \to \langle \vec{p} \rangle$$

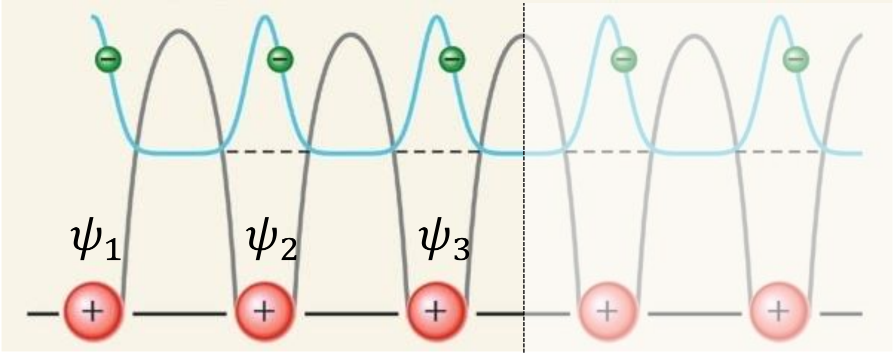
$$m_e \to m(\vec{p})$$

Quantum mechanics

$$i\hbar \frac{d|\Psi(\mathrm{t})\rangle}{dt} = H|\Psi(\mathrm{t})\rangle$$

$$|\Psi\rangle = \begin{pmatrix} \psi_0 \\ \psi_1 \\ \psi_2 \end{pmatrix}$$

$$H = \begin{pmatrix} \epsilon_0 & t_{01} & 0 \\ t_{10} & \epsilon_1 & t_{12} \\ 0 & t_{21} & \epsilon_2 \end{pmatrix}$$



$$\begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} \quad \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \quad \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}$$

The Hamiltonian as Sparse matrix

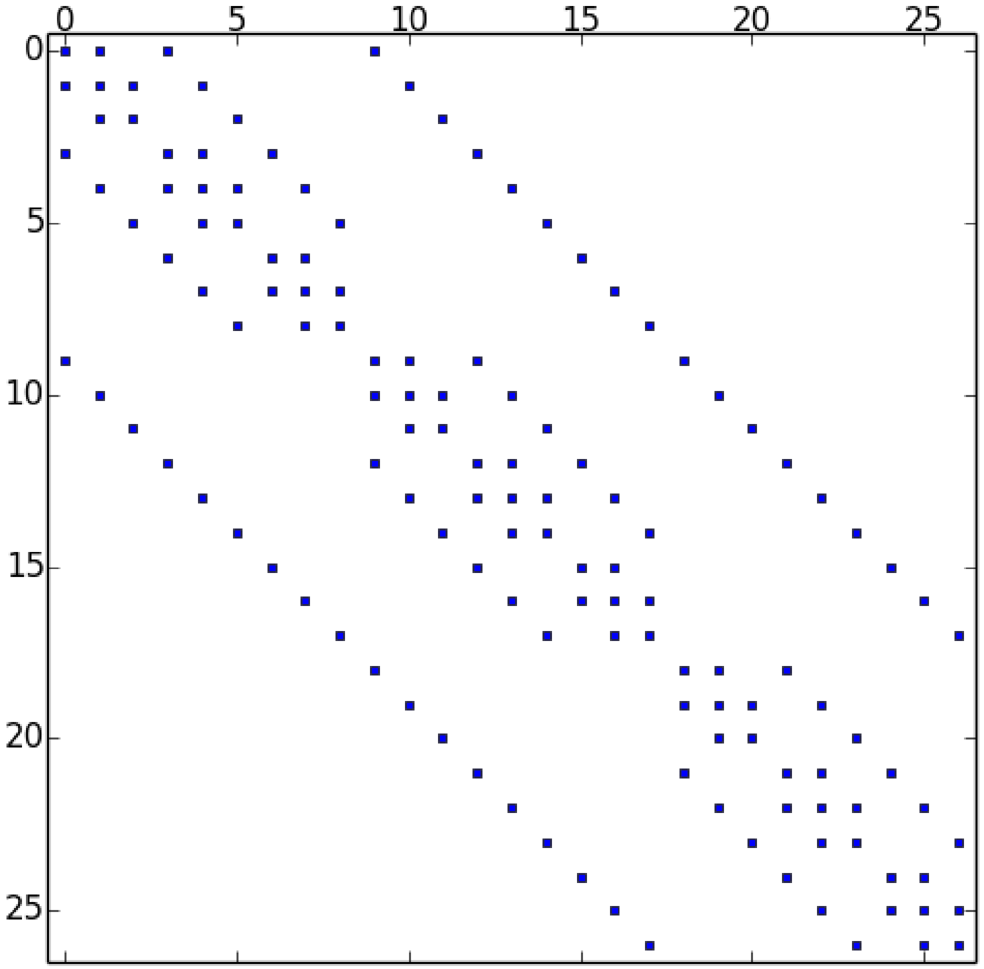
$$H = \begin{pmatrix} \epsilon_0 & \cdots & 0 \\ \vdots & \ddots & \vdots \\ 0 & \cdots & \epsilon_d \end{pmatrix}$$

$d \times d$ elements

Column	Row	Matrix element

$$\begin{pmatrix} 0 \\ 0 \\ 1 \\ \vdots \\ d \end{pmatrix} \quad \begin{pmatrix} 0 \\ 1 \\ 1 \\ \vdots \\ d \end{pmatrix} \quad \begin{pmatrix} \epsilon_0 \\ t_{01} \\ \epsilon_1 \\ \vdots \\ \epsilon_d \end{pmatrix}$$

$3\ d$ elements



The macroscopic *A*

$$i\hbar \frac{d|\Psi(\mathrm{t})\rangle}{dt} = H|\Psi(\mathrm{t})\rangle$$

$$\langle \hat{A}(t)\rangle = \langle \Psi(t)|\hat{A}|\Psi(\mathrm{t})\rangle \equiv \mathrm{Tr}[\hat{A}\rho(t)]$$ Density matrix

$$i\hbar \frac{d\rho}{dt} = [H,\rho]$$

Von Neumann equation

The electrical response of *A*

$$\langle \hat{A}(t, \vec{E}\,) \rangle \equiv \mathrm{Tr}[\hat{A}\,\rho(H, \vec{E})]$$

$\vec{E} = -\nabla\phi(\vec{r})$

$$H = H_0 + \int d^3r\, n(\vec{r})\phi(\vec{r})$$

Disordered Hamiltonian

Minimal coupling

The electrical response of *A*

$$\langle \hat{A}(t, \vec{E}\,) \rangle \equiv \mathrm{Tr}[\hat{A}\,\rho(H, \vec{E})]$$

$$\vec{E} = -\nabla\phi(\vec{r})$$

$$H = H_0 + \int d^3r\, n(\vec{r})\phi(\vec{r})$$

Disordered Hamiltonian

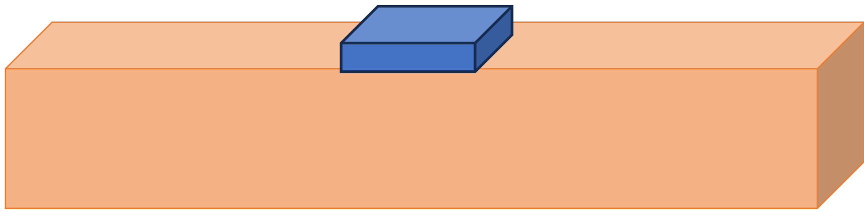
Minimal coupling

Strong electric fields could change the ground states!

Linear response theory: The Kubo formula

Principal goal:

$$\rho\left(H,\vec{E}\right) = \rho_0(H) + \left(\nabla_{\vec{E}}\,\rho\left(\vec{E},H\right)\right)_{\vec{E}=0}\vec{E}$$



Fundamental Hypotheses

- Thermal equilibrium: $\rho_0(H) \rightarrow \rho_{\mathrm{eq}}(\mathrm{T},\mu)$

Linear response theory

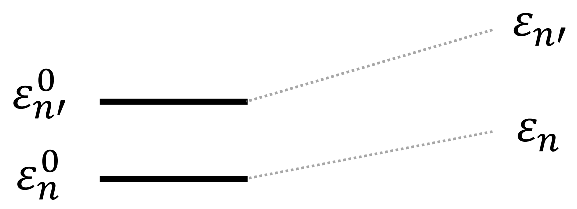
Principal goal:

$\chi_{\hat{E}}$

$$\rho\left(H,\vec{E}\right) = \rho_0(H) + \left(\nabla_{\vec{E}}\,\rho\left(\vec{E},H\right)\right)_{\vec{E}=0}\vec{E}$$

Fundamental Hypotheses

• Thermal equilibrium:	$\rho_0(H) \to \rho_{eq}(\mathrm{T},\mu)$
• Adiabatic evolution:	$\lvert\varepsilon_n^0\rangle \to \lvert\varepsilon_n(t)\rangle$



$$\langle\hat{A}(t)\rangle \equiv \mathrm{Tr}[\hat{A}(t)\rho_{eq}] + \mathrm{Tr}[\hat{A}(t)\chi_{\hat{E}}]$$

Linear response theory: The Kubo formula

$$\langle \hat{A}(t) \rangle = \lim_{t,\tau\to\infty} \int_0^{\beta} d\beta' \int_0^{t} d\tau' e^{-\tau'/\tau} \langle \rho_{eq}\, \hat{A}\, [E_\alpha J_\alpha(\tau' - i\hbar\beta')] \rangle$$

- ρ_{eq}: Density matrix in thermal equilibrium
- $E_\alpha J_\alpha(\tau' - i\hbar\beta')$: **History** of the electric field coupled to the current density given a thermal broadening

Kubo, Ryogo (1957). "Statistical-Mechanical Theory of Irreversible Processes. I. General Theory and Simple Applications to Magnetic and Conduction Problems". J. Phys. Soc. Jpn. 12 (6): 570–586. doi:10.1143/JPSJ.12.570.º

Noninteracting Kubo Formula

$$\langle \hat{A} \rangle = i\hbar \int dE \, \mathrm{Tr}\left[f(E' - \mu)\delta(E' - H)\widehat{\mathrm{A}} \, \frac{\mathrm{d\,G^+(E')}}{dE} \vec{V} \cdot \vec{E}_0 \right]$$

Noninteracting Kubo Formula

$$\langle \hat{A} \rangle = i\hbar \int dE \, \mathrm{Tr}\left[f(E' - \mu)\delta(E' - H)\hat{\mathrm{A}} \, \frac{\mathrm{d\,G^{+}(E')}}{dE} \vec{V} \cdot \vec{E}_0 \right]$$

Representable in tight-binding basis

$$H = \begin{pmatrix} \epsilon_0 & t_{01} & 0 \\ t_{10} & \epsilon_1 & t_{12} \\ 0 & t_{21} & \epsilon_2 \end{pmatrix} \quad |\Psi\rangle = \begin{pmatrix} \psi_0 \\ \psi_1 \\ \psi_2 \end{pmatrix}$$

Noninteracting Kubo Formula

$$\langle \hat{A} \rangle = i\hbar \int dE \, \mathrm{Tr}\left[f(E' - \mu)\delta(E' - H)\hat{\mathrm{A}} \frac{\mathrm{d\,G^{+}(E')}}{dE} \vec{V} \cdot \vec{E}_0 \right]$$

Representable in tight-binding basis

$$H = \begin{pmatrix} \epsilon_0 & t_{01} & 0 \\ t_{10} & \epsilon_1 & t_{12} \\ 0 & t_{21} & \epsilon_2 \end{pmatrix} \quad |\Psi\rangle = \begin{pmatrix} \psi_0 \\ \psi_1 \\ \psi_2 \end{pmatrix}$$

$$\langle \psi_n | V_x | \psi_m \rangle = i(X_n - X_m) H_{nm}$$

Noninteracting Kubo Formula

$$\langle \hat{A} \rangle = i\hbar \int dE \, \mathrm{Tr}\left[f(E' - \mu)\delta(E' - H)\hat{\mathrm{A}} \, \frac{\mathrm{d\,G^+(E')}}{dE} \vec{V} \cdot \vec{E}_0 \right]$$

Singular Functions

$$\mathrm{G^+(E')} = \lim_{\eta \to 0} \frac{1}{H - E + i\eta}$$

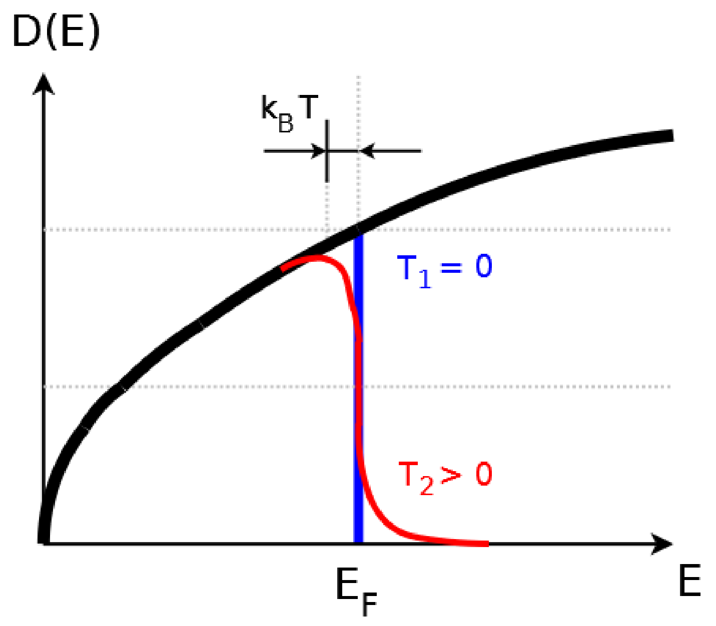
$$\int dE' \delta(\mathrm{E'} - H) f(E') = f(H)$$

Numerical implementation

The kernel polynomial method

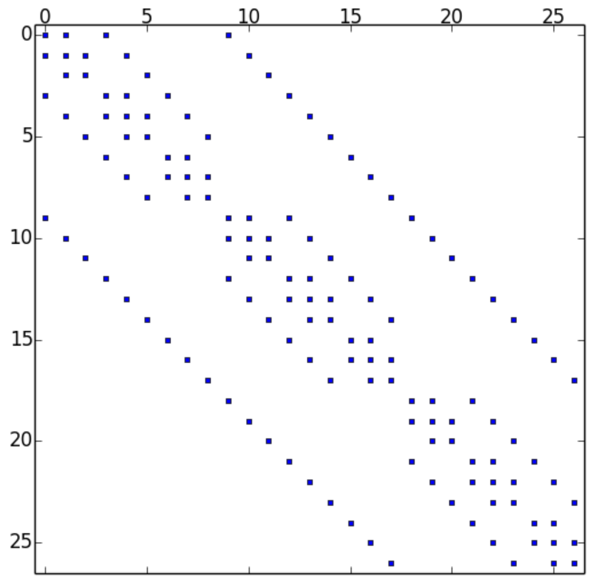
$$DOS \equiv \mathrm{Tr}[\delta(\mathrm{E}' - H)]$$

$$Tr[\delta(H - E)] = \sum_n \delta(E_n - E)$$



Polynomial expansion

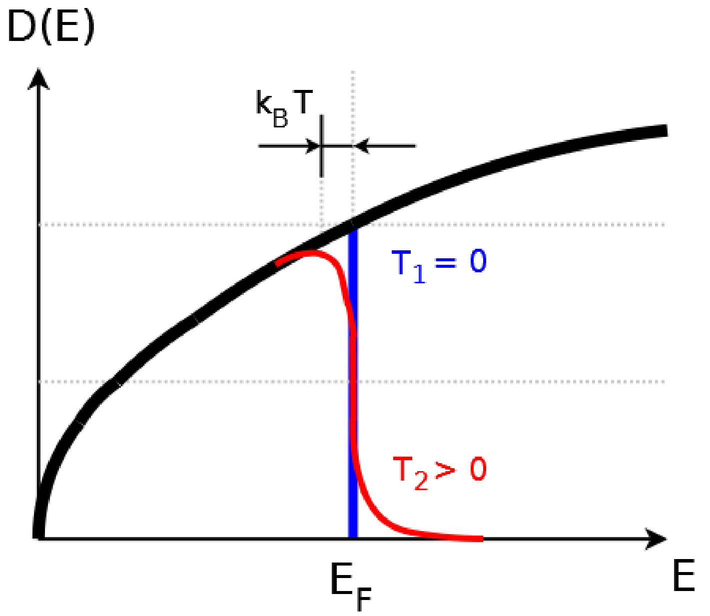
$$\delta(H - E) = \sum_{n=0}^{\infty} c_n \, p_n(H)$$



The kernel polynomial method

$$DOS \equiv \mathrm{Tr}[\delta(\mathrm{E}' - H)]$$

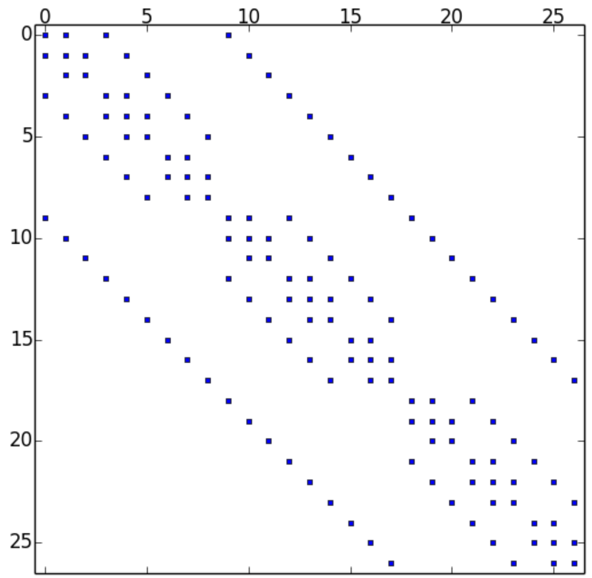
$$Tr[\delta(H - E)] = \sum_n \delta(E_n - E)$$



Polynomial expansion

$$\delta(H - E) = \sum_{n=0}^{\infty} c_n \, p_n(H)$$

$$O(N^3) \rightarrow O(N^2)$$



The random phase approximation

$$DOS \equiv \mathrm{Tr}[\delta(\mathrm{E}' - H)]$$

$$DOS = \langle r|\delta(H - E)|r\rangle$$

$$= \sum_{n,m=0}^{d} \xi_n \, \xi_m \langle n|\delta(H - E)|m\rangle$$

$$|r\rangle = \begin{pmatrix} \xi_0 \\ \vdots \\ \xi_d \end{pmatrix}$$

$$O(N^2) \to O(N)$$

Choosing the right distribution

$\xi_n \, \xi_m = 0$

$\xi_n^2 = 1$

The Chebyshev polynomials

$$\mathrm{Tr}[\delta(H - E)] = \sum_{n=0}^{M} c_n \langle r | p_n(H) | r \rangle$$

$$T_m(x) \equiv Cos\left(m\, ArcCos\,(x)\right)$$

$$T_{n+1}(x) \equiv 2\,T_n(x) - T_{n-1}(x)$$

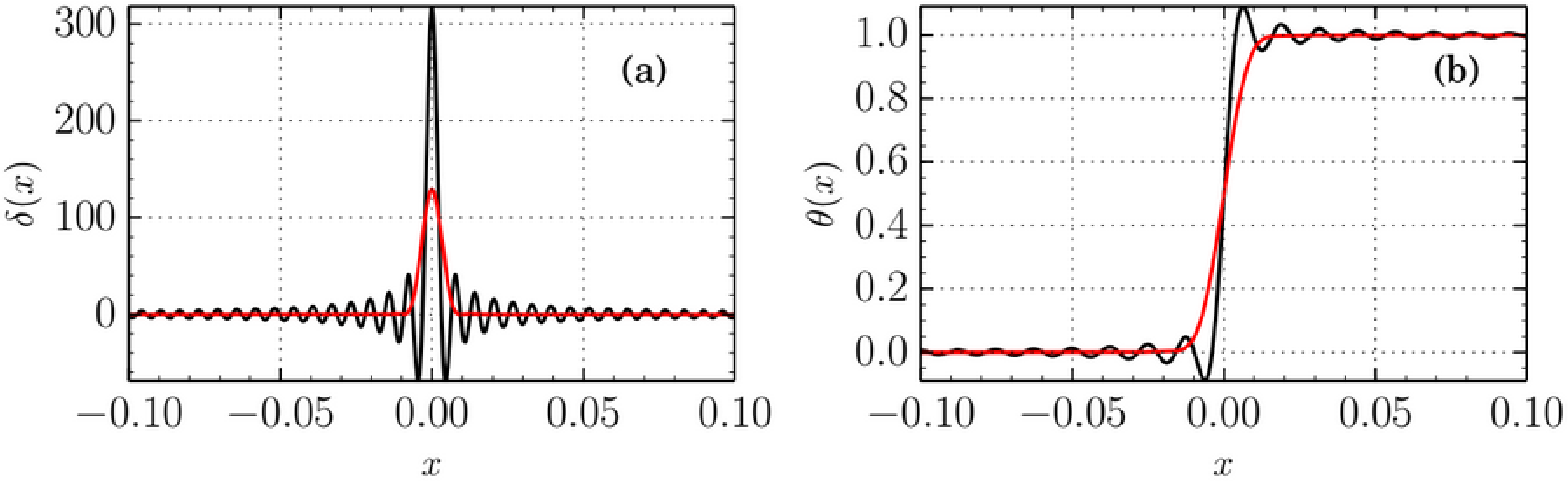
$$T_0(x) \equiv 1,$$
$$T_1(x) \equiv x\ ,$$
$$T_2(x) \equiv 2x^2 - 1,$$
$$T_3(x) \equiv 4x^3 - 2x$$

$$O\left(N^2\right) \to O(M\,N)$$

The Gibbs oscillations

$$\delta(x - x_0) = \frac{2}{\pi\sqrt{1 - x^2}} \sum_{m=0}^{M} \frac{g_m \mathrm{T}_m(x_0)\mathrm{T}_m(x)}{1 + \delta_{m0}}$$

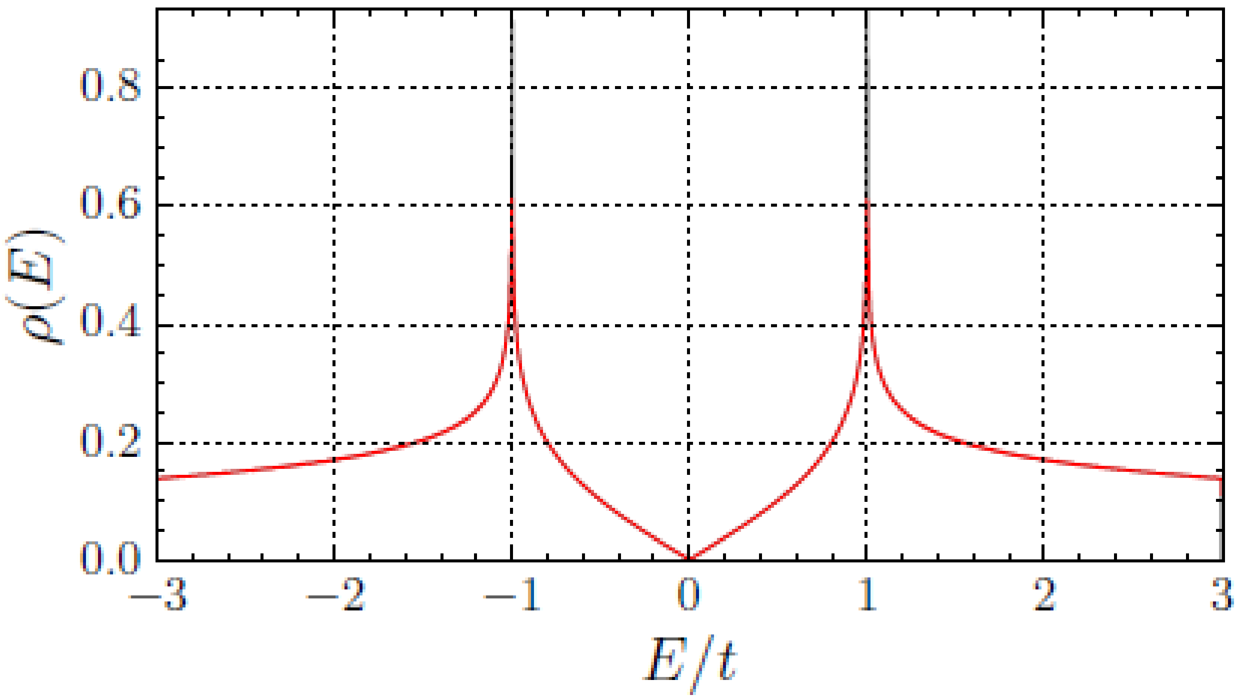
$$g_m = \frac{\sinh\left(\left(1 - \frac{m}{M}\right)\lambda\right)}{\sinh(\lambda)}$$



The final density of states

$$\mathrm{DOS} = \frac{2\alpha}{W_H}\frac{2}{\pi\sqrt{1-\tilde{\varepsilon}_F}}\sum_{m=0}^{M} g_m \mathrm{T}_m(\tilde{\varepsilon}_F)\mu_m$$

$$\mu_m = \mathrm{Tr}\left[\,\mathrm{T}_m(\widetilde{H})\right] = \langle\langle\ \langle r|\mathrm{T}_m(\widetilde{H})|\mathrm{r}\rangle\ \rangle\rangle$$

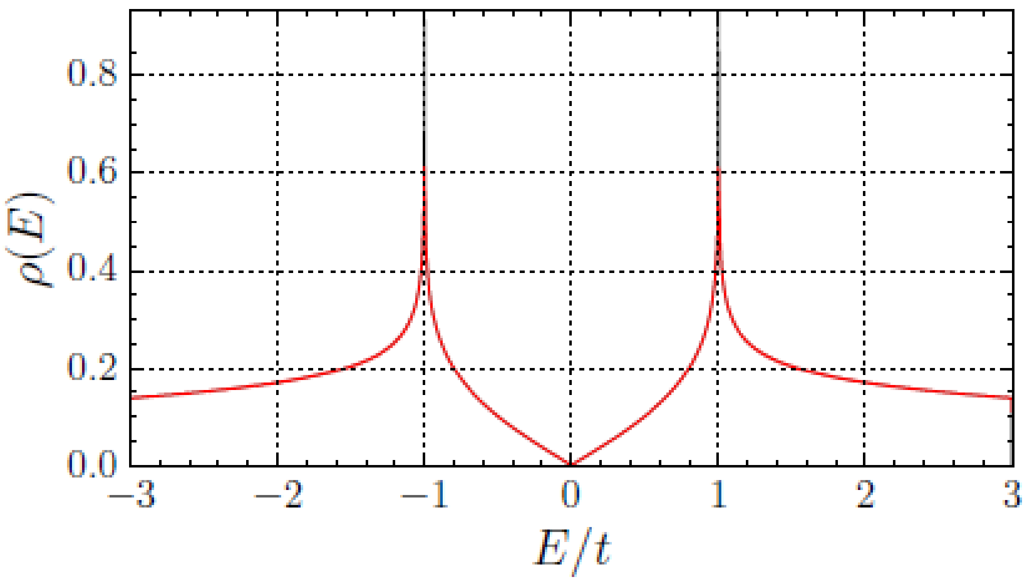


$$\widetilde{H}, \widetilde{\varepsilon}_F \in (-\alpha, \alpha) \quad \widetilde{H} = \frac{2\alpha}{W_H}(\,H - E_c), \widetilde{\varepsilon}_F = \frac{2\alpha}{W_H}(\,\varepsilon_F - E_c)$$

The final density of states

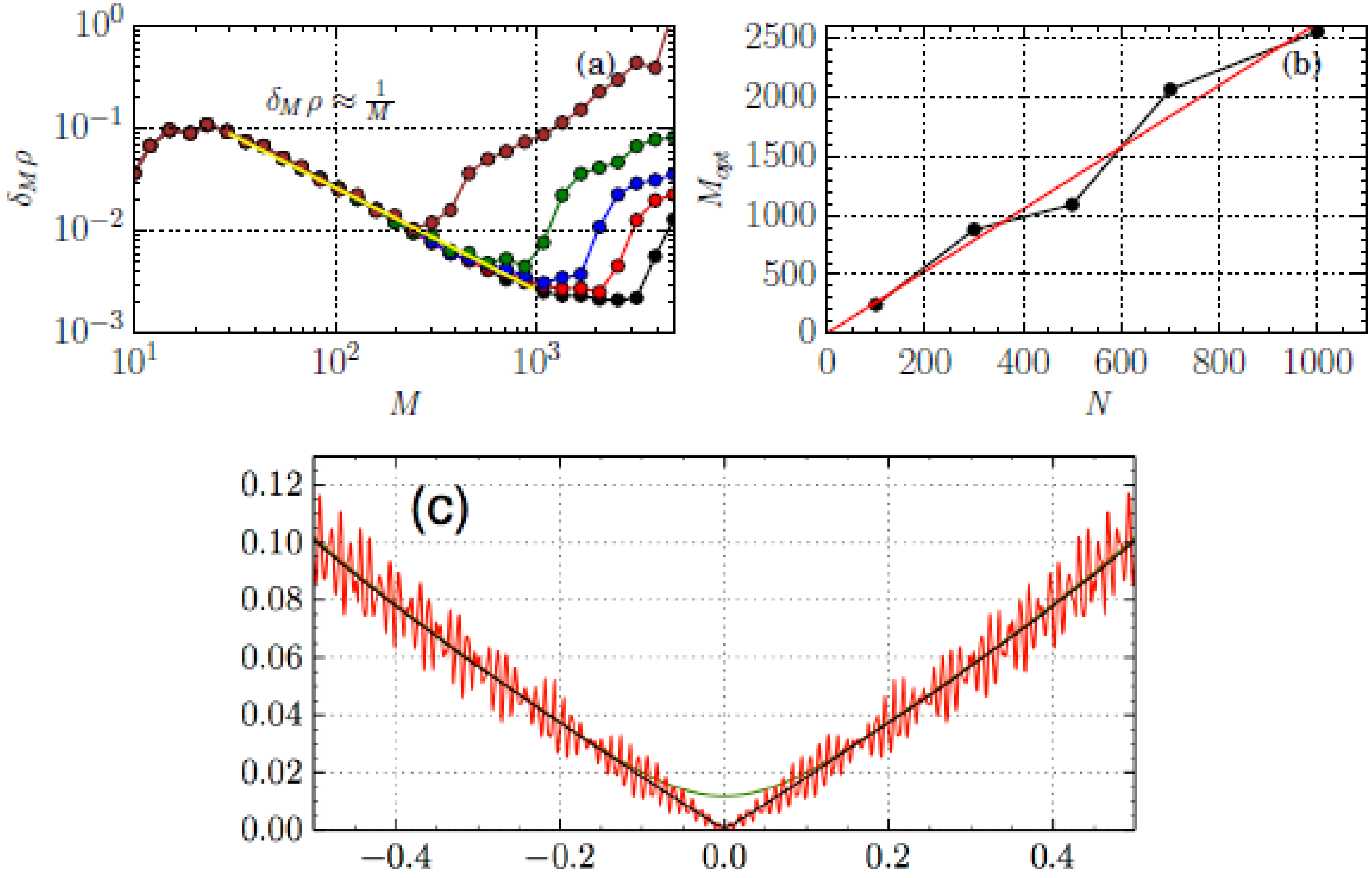
The density of states using KQuant

```
import matplotlib.pyplot as plt
from k_quant.system import System

import kpm
kpm.safe_CUTOFF= 0.99;

wann_syst = System( dimensions = (10000,1,1), w90_inp="linear_chain")
dens = kpm.Density(wann_syst, bounds=(-1,1))
dens.ComputeMoments( broadening =0.05)

plt.plot(*dens.spectral_average() )
```



https://www.lsquant.org/lsq/index.php

Convergence of the Chebyshev polynomials



The LSQT

$$\langle \hat{A} \rangle = i\hbar \int dE \, \mathrm{Tr}\left[f(E' - \mu)\delta(E' - H)\hat{\mathrm{A}} \, \frac{\mathrm{d\,G^{+}(E')}}{dE} \vec{V} \cdot \vec{E}_0 \right]$$

$$\mu_{m,n}$$

$$\langle \hat{A}(\mu, T) \rangle = \sum_{m=0}^{M} \sum_{n=0}^{M} \Gamma_{m,n}(\mu, T) \, \mathrm{Tr}\left[\mathrm{T}_m(H) \hat{A} \mathrm{T}_n(H) \hat{V}_\beta \right]$$

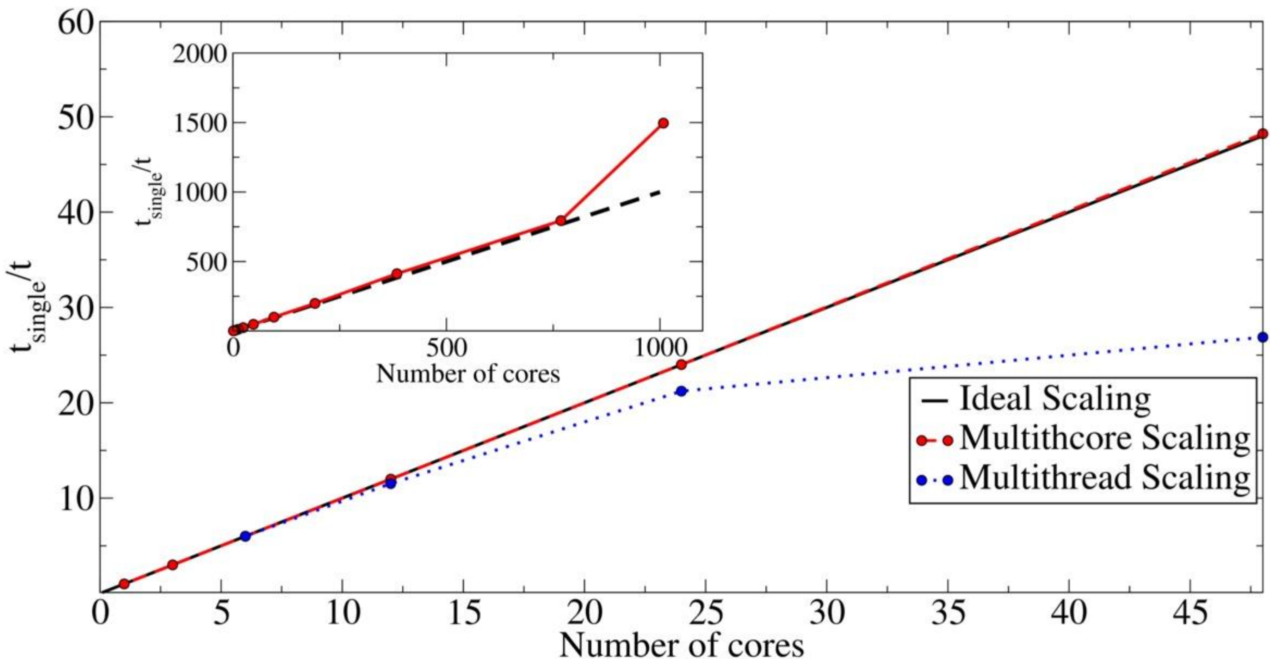
Chebyshev expansion moment matrix

Scaling of the LSQT

$$\mu_{m,n} = \mathrm{Tr}\left[T_m(H) j_x T_n(H) j_x\right]$$

μ_{00}	$\mu_{0,1}$	$\mu_{0,2}$	$\cdots$	$\mu_{0,M-1}$	$\mu_{0,M}$	Process #1
$\mu_{1,0}$	$\mu_{1,1}$	$\mu_{1,2}$	$\cdots$	$\mu_{1,M-1}$	$\mu_{1,M}$	Process #2
$\mu_{2,0}$	$\mu_{2,1}$	$\mu_{2,2}$	$\cdots$	$\mu_{2,M-1}$	$\mu_{2,M}$	Process #3
$\vdots$	$\vdots$	$\vdots$	$\ddots$	$\vdots$	$\vdots$	
$\mu_{M-1,0}$	$\mu_{M-1,1}$	$\mu_{M-1,2}$	$\cdots$	$\mu_{M-1,M-1}$	$\mu_{M-1,Ms}$	Process #N-1
$\mu_{M,0}$	$\mu_{M,1}$	$\mu_{M,2}$	$\cdots$	$\mu_{M,M-1}$	$\mu_{M,M}$	Process #N

Multi-threaded sparse matrix-vector multiplication

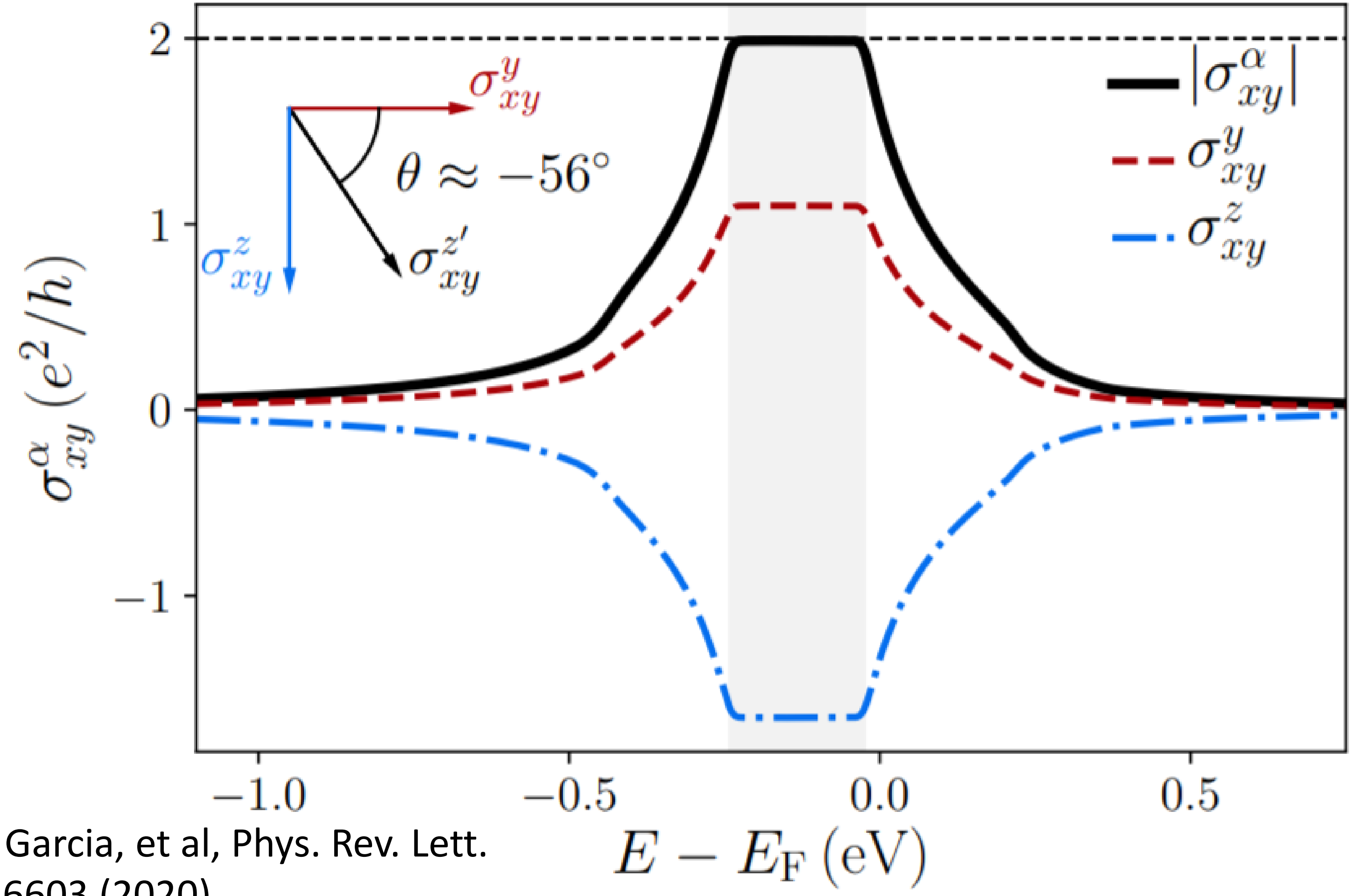


Aplications

Applications: Canted quantum Spin Hall effect

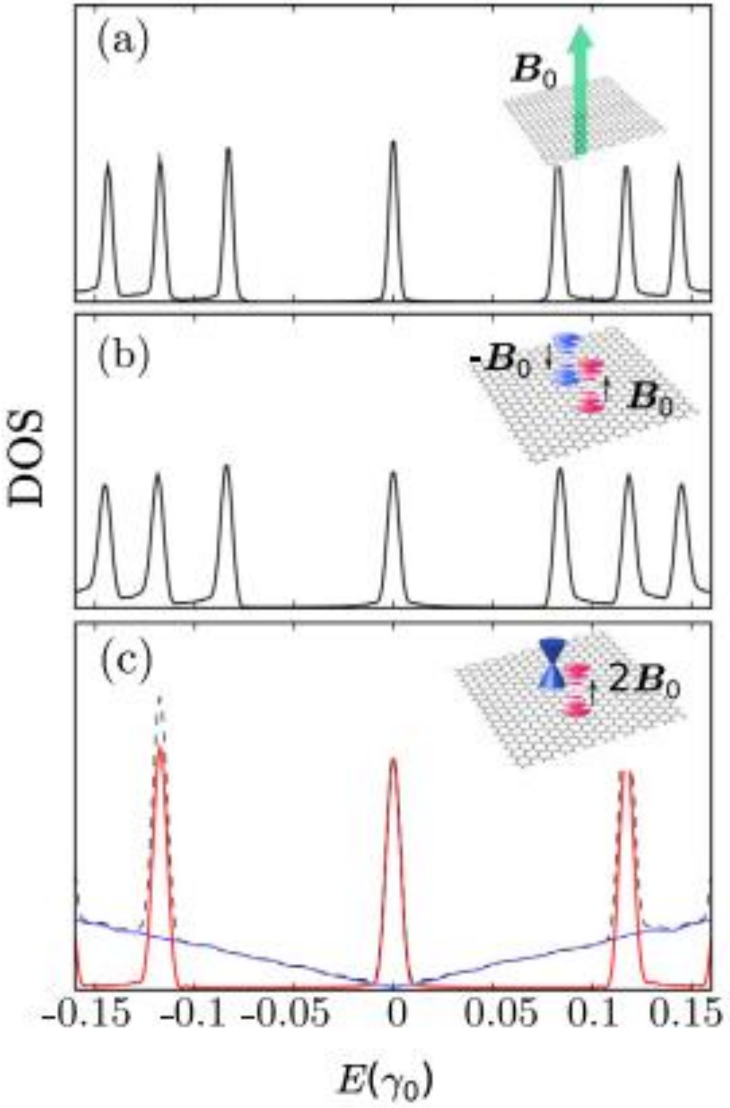
$\langle \vec{s} \rangle_{E_F} \approx \langle S_0 \rangle_{E_F}\, \hat{u}(\theta)$

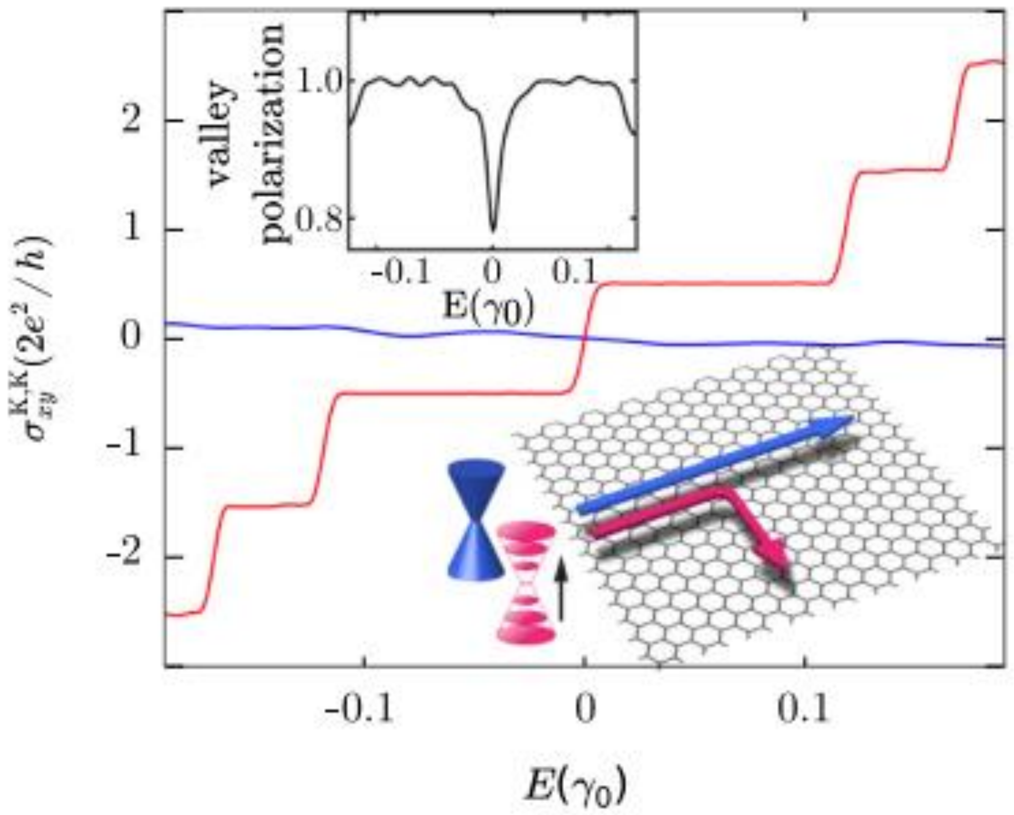
$\theta \equiv \arctan\left(\Lambda_z/\Lambda_y\right) \approx -56°$



Jose H. Garcia, et al, Phys. Rev. Lett. 125, 256603 (2020)

Applications: Valley Hall effect

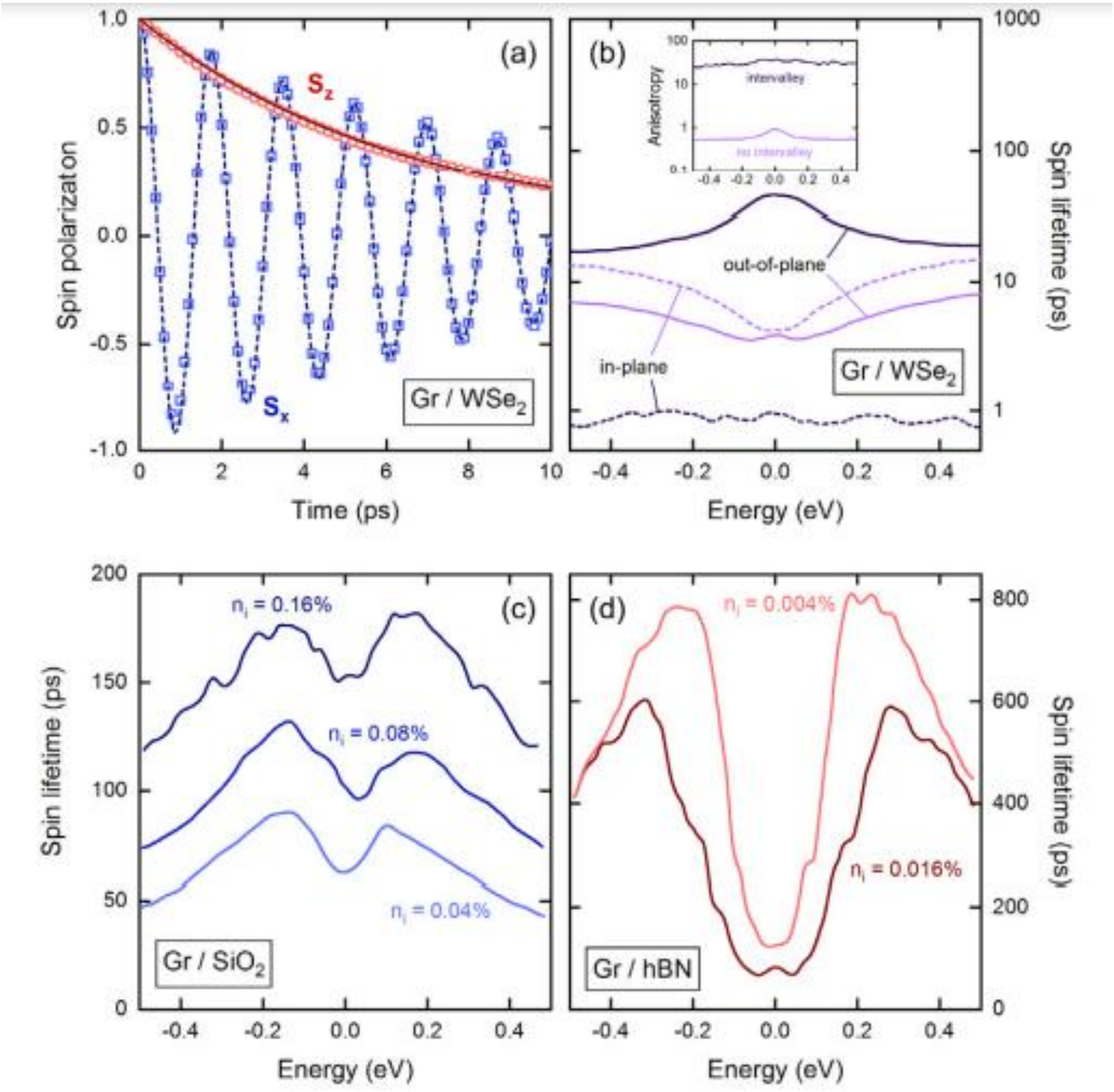




$$\hat{J}_{\alpha}^{v} = \frac{1}{2}(P_v^{+}\hat{J}_{\alpha}P_v^{+} - P_v^{-}\hat{J}_{\alpha}P_v^{-}).$$

$$P_v^{\pm} = \sum_{\boldsymbol{k}} \theta(|\boldsymbol{k} - \boldsymbol{K}^{\pm}| - R)|\boldsymbol{k}\rangle\langle\boldsymbol{k}|$$

Applications: Spin relaxation



$$\mathbf{S}(E,t) = \frac{1}{2}\frac{\langle\phi(t)|\,\hat{\mathbf{s}}\delta(E-\hat{H})\,|\phi(t)\rangle + \text{h.c.}}{\langle\phi(t)|\,\delta(E-\hat{H})\,|\phi(t)\rangle},$$

European Research Council
Established by the European Commission

GRAPHENE FLAGSHIP

Jaime Garrido
Automatic parameter optimization

Joaquin Medina
Adaptation to spin-orbit torque

Luis Canonico
Implementation of momentum-efficient KPM

Josep Buch
Exploration of machine-learning Hamiltonian models