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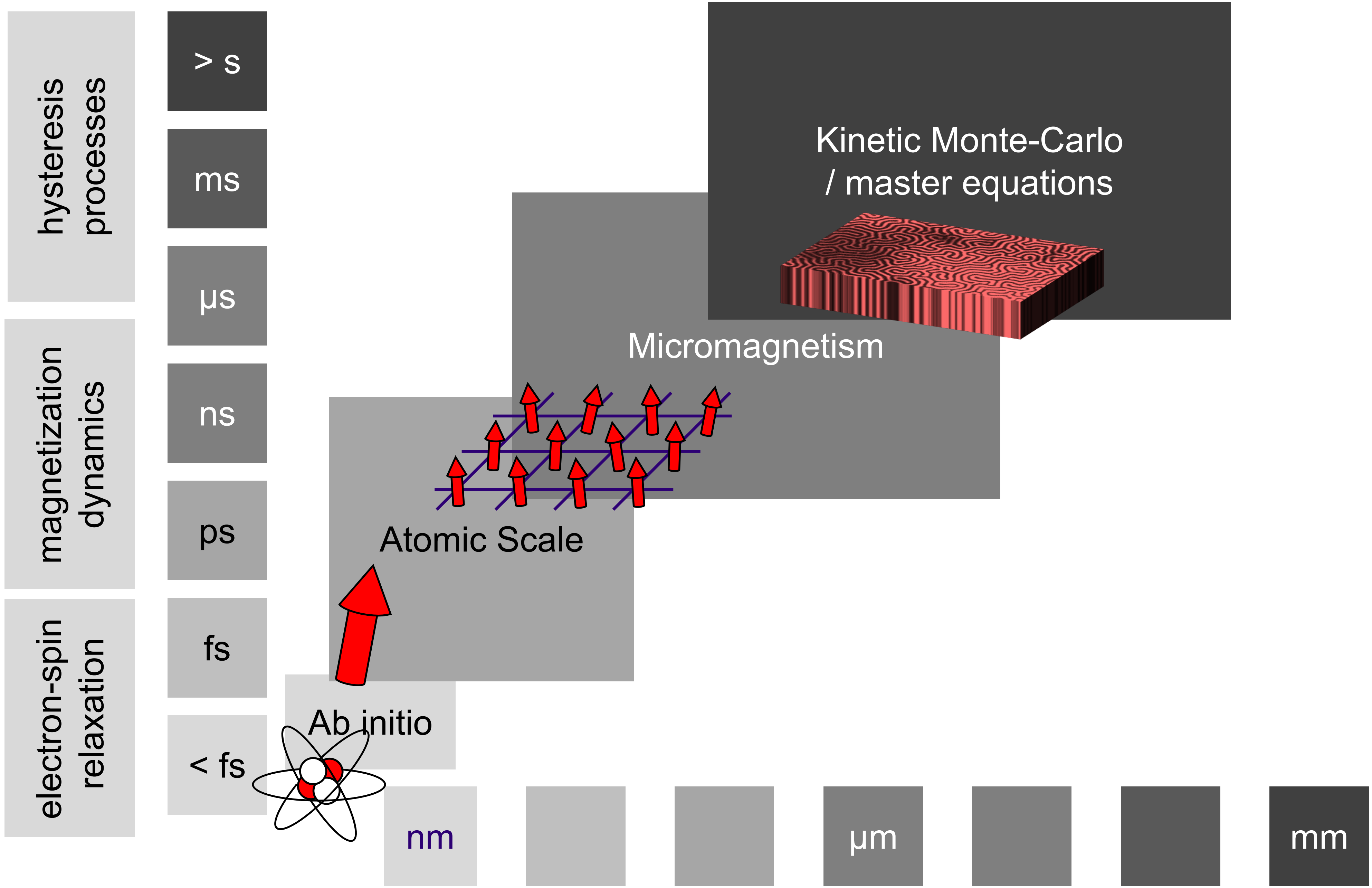
Atomistic spin dynamics with a quantum thermostat

Joseph Barker

University of Leeds

Different approaches for different scales



Multiscale modelling

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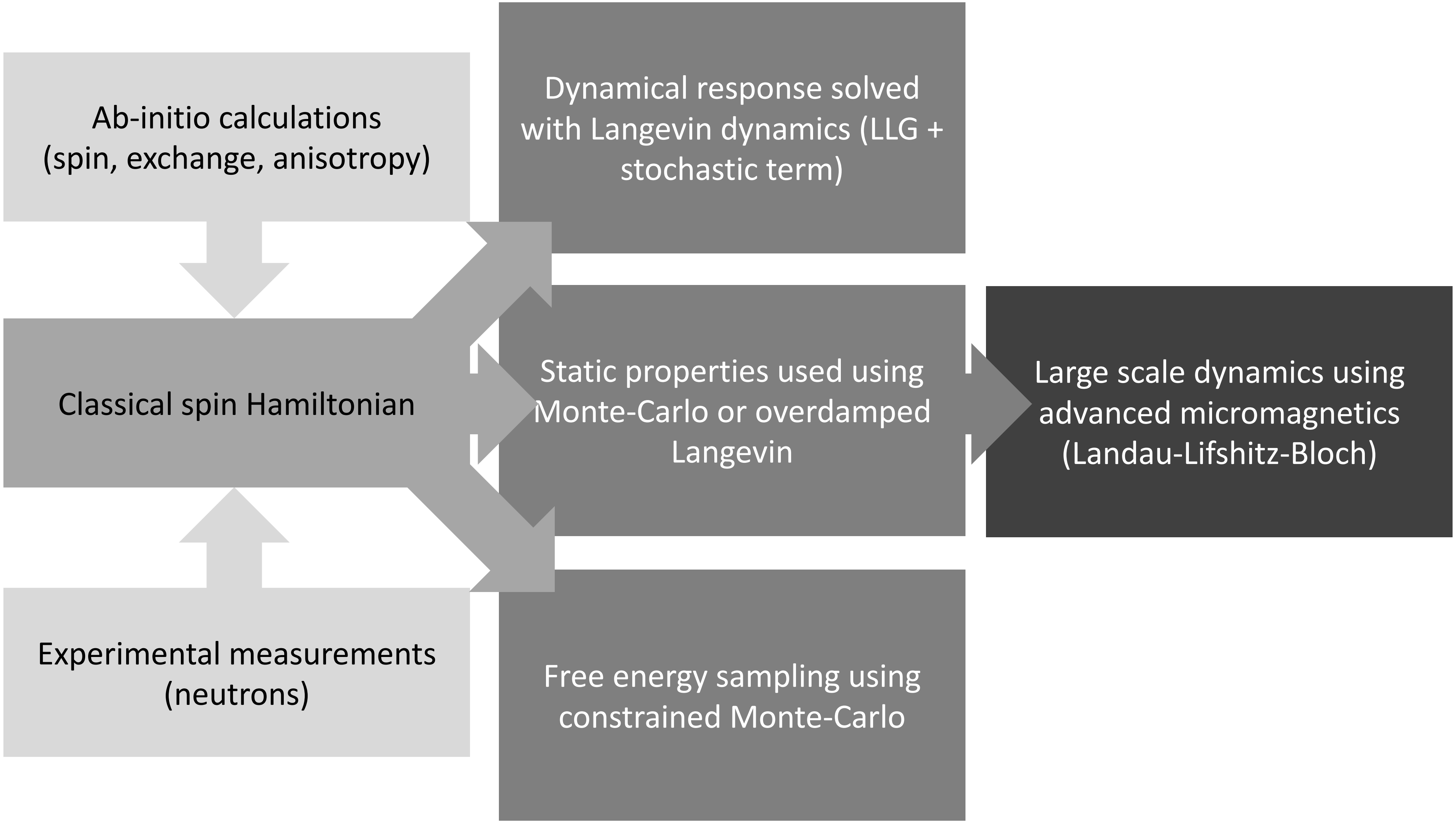

Atomistic spin dynamics (ASD)

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Heisenberg Hamiltonian

Classical spin model: **S** is a unit vector

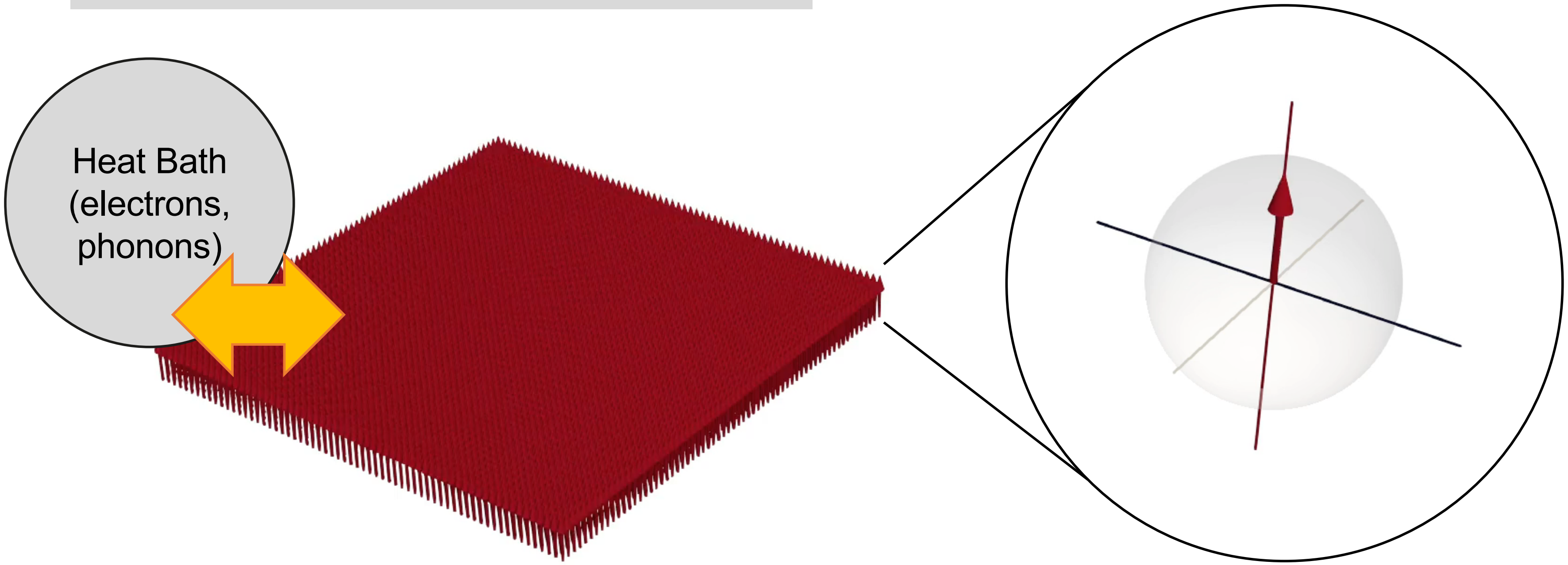
$$\mathcal{H} = -\sum_{ij} \mathbf{S}_i \mathbb{J}_{ij} \mathbf{S}_j$$

Landau-Lifshitz-Gilbert

Equation of motion for a classical spin in a local field.

$$\frac{\partial \mathbf{S}_i}{\partial t} = -\frac{\gamma_i}{(1+\alpha_i^2)} \left(\mathbf{S}_i \times \mathbf{H}_i + \alpha_i \mathbf{S}_i \times \mathbf{S}_i \times \mathbf{H}_i \right)$$

Simplest spin-spin Hamiltonian
Many more terms can be included



Inclusion of the heat bath allows thermodynamic calculations

ASD – Heisenberg Hamiltonian

Heisenberg Hamiltonian

Classical spin model: **S** is a unit vector

$$\mathcal{H} = -\sum_{ij} \mathbf{S}_i \mathbb{J}_{ij} \mathbf{S}_j$$

$$\mathbb{J} = \begin{pmatrix} J_{xx} & J_{xy} & J_{xz} \\ J_{yx} & J_{yy} & J_{yz} \\ J_{zx} & J_{zy} & J_{zz} \end{pmatrix}$$

Ab initio parameterization means this can be long ranged.

Anisotropic Exchange

$$J_{xx} = J_{yy} \neq J_{zz}$$

Example: two-ion exchange in FePt

J. Barker et al. APL 97, 192504 (2010)

Dzyaloshinskii-Moria Interaction

$$\mathcal{H}_{\mathrm{DM}} = \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$$

Example: antiferromagnetic Skyrmions

J. Barker et al. arXiv:1505.06156 [cond-mat.mes-hall]

ASD - Additional Terms

Magneto-Crystalline Anisotropy

Temperature dependence of anisotropy is important for applications. Can also cause reorientation transitions.

$$\mathcal{H}_{\mathrm{MCA}} = \frac{\kappa_2}{2}\left(3S_z^2 - 1\right) + \frac{\kappa_4}{8}\left(35S_z^4 - 30S_z^2 + 3\right) + \frac{\kappa_6}{16}\left(231S_z^6 - 315S_z^4 + 105S_z^2 - 5\right)$$

Dipole-dipole interactions

Computationally expensive and usually more relevant on a micromagnetic scale. Can be important to calculate in non-cubic systems.

$$\mathcal{H}_{\mathrm{dipole}} = -\frac{1}{2}\left(\frac{\mu_s^2\mu_0}{4\pi a^3}\right)\sum_{ij}\frac{3(\mathbf{S}_i\cdot\hat{\mathbf{e}}_{ij})(\hat{\mathbf{e}}_{ij}\cdot\mathbf{S}_j) - \mathbf{S}_i\cdot\mathbf{S}_j}{r_{ij}^3}$$

Atomistic spin dynamics

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Temperature
Phase transitions
Thermal stability
Non-equilibrium
Quantum statistics

Spin waves
Intrinsic damping
Temperature dependence

ATOMISTIC SPIN DYNAMICS

Disorder
Spin wave scattering
Amorphous materials
Rough interfaces
Impurities

Multiscale
Realistic unit cell
Complex materials
Parameterization from ab initio

When is a GPU faster?

The speed depends on the algorithm and the memory access requirements.

Vector processor

Same operation on large arrays
Large memory bus
Acts as a coprocessor
Branching is bad

Optimized for floats

Fast intrinsic functions
Integer ops can be slow

Not just a drop in accelerator!

GPUS – Thread/memory arrangement

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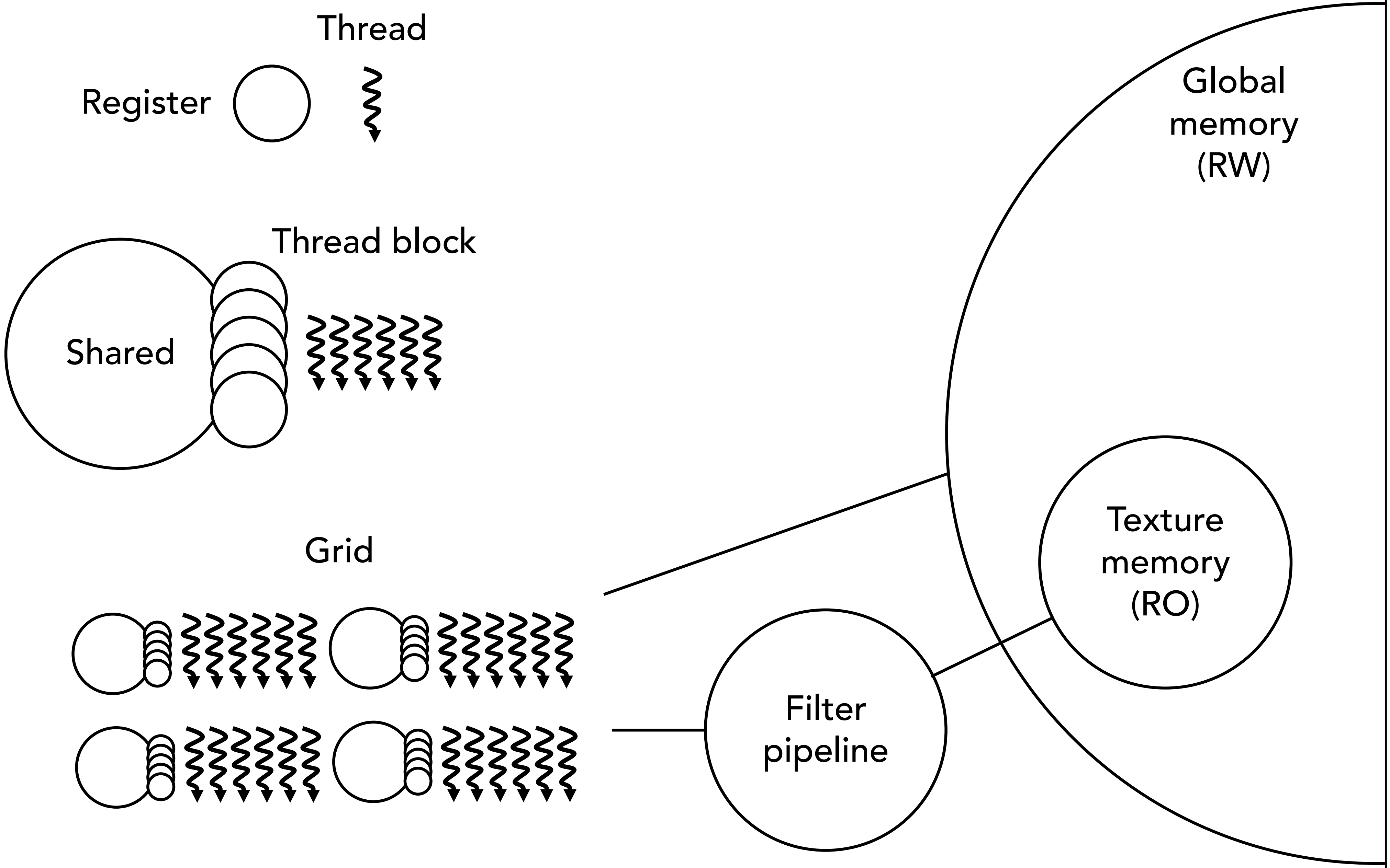

GPUs – Host / Device relationship

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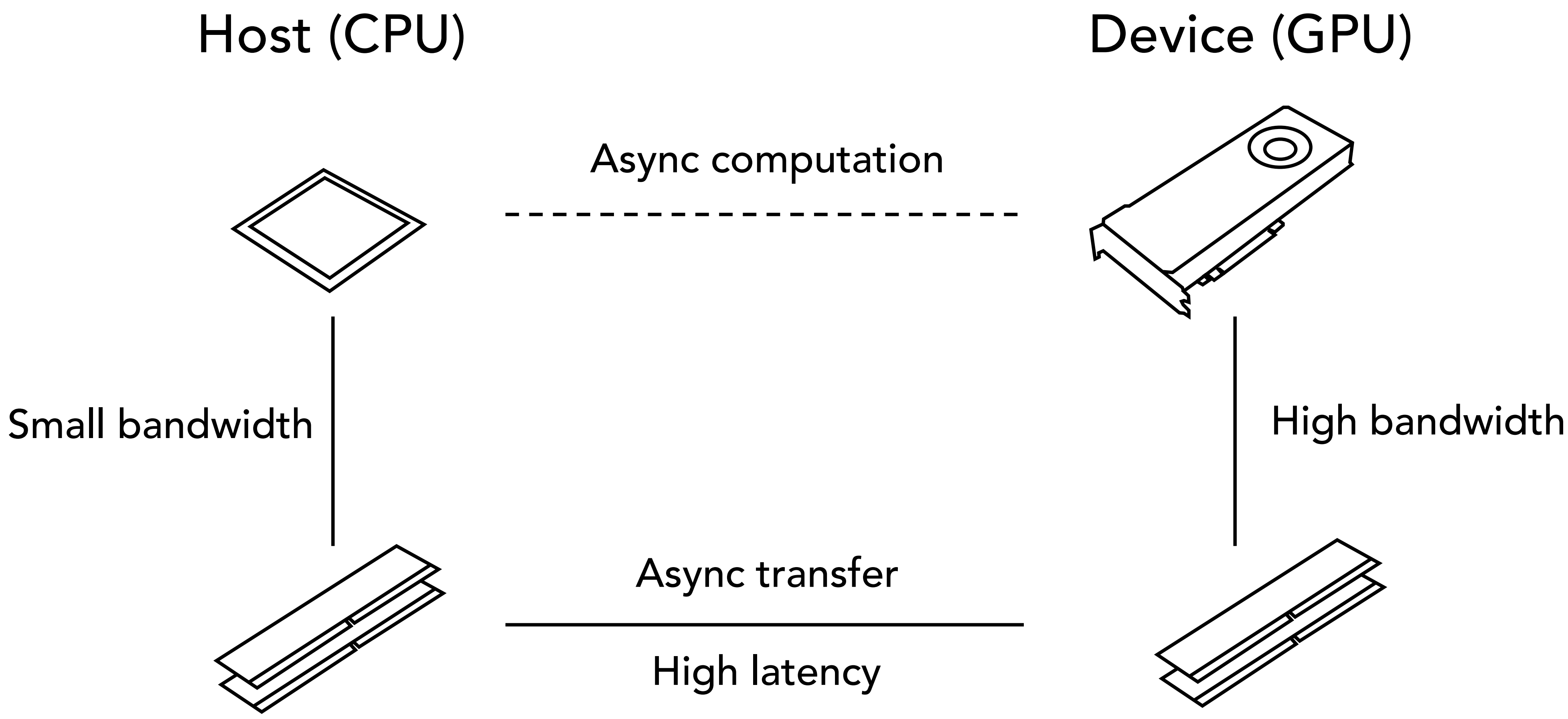

GPUS – Exchange calculation

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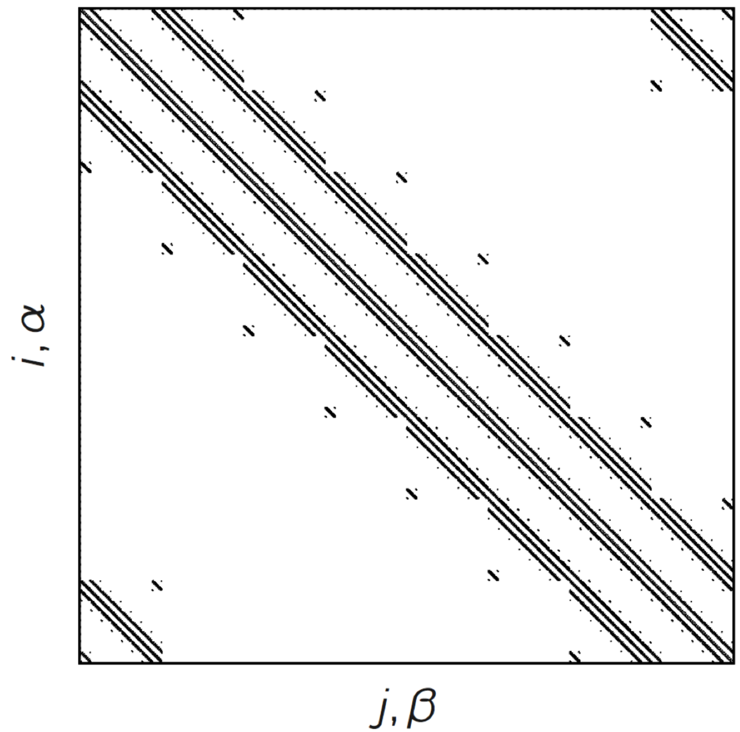
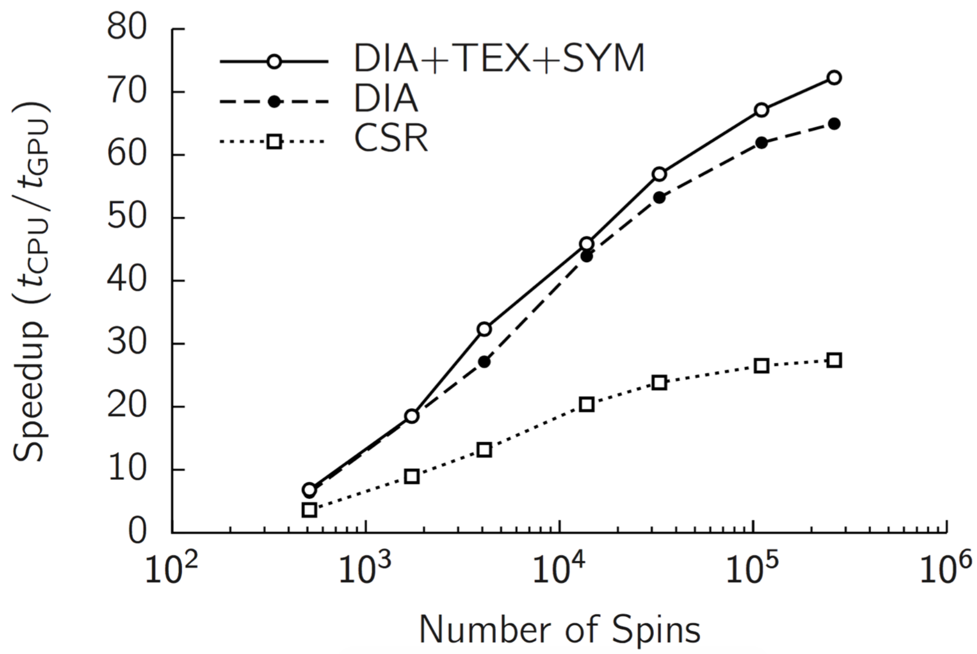
Different storage strategies

Large performance difference exist depending on how data is stored and the algorithms used.





GPUS – Parallel streams

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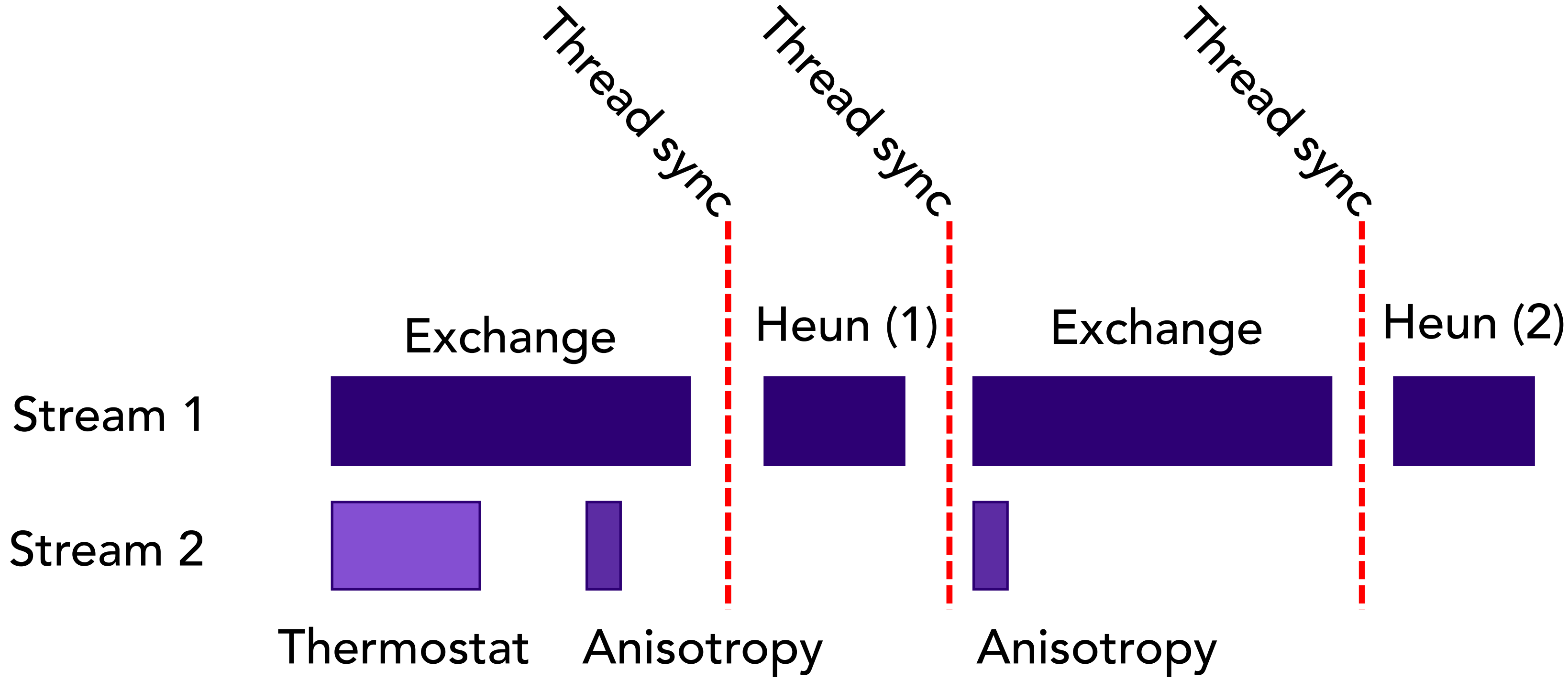
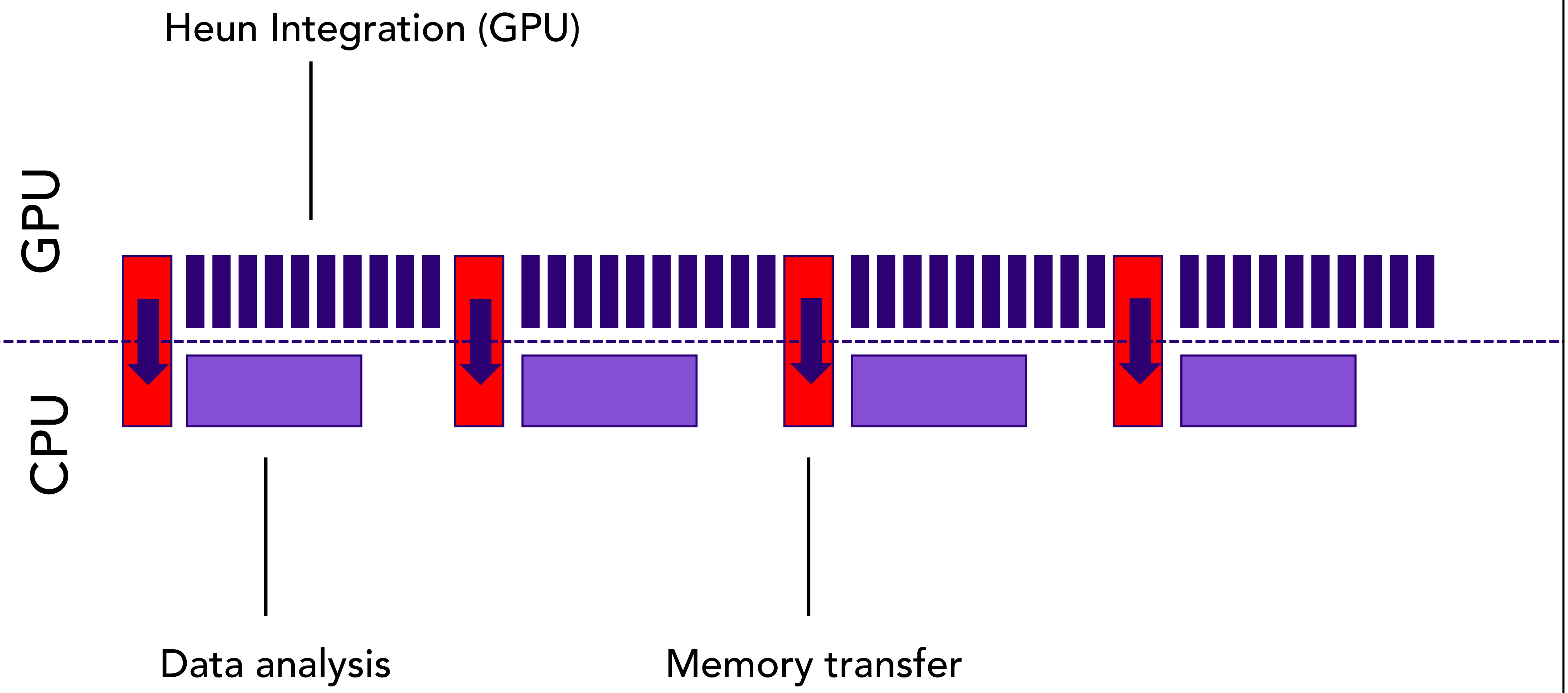
Memory access can be hidden with streams

Overlapping computation keeps the multiprocessors busy while some kernels are waiting for memory access operations to complete.



GPUS – Coprocessing



Atomistic Spin Dynamics

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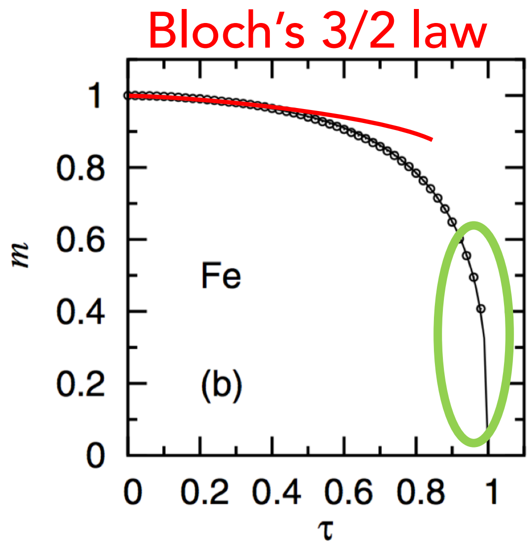
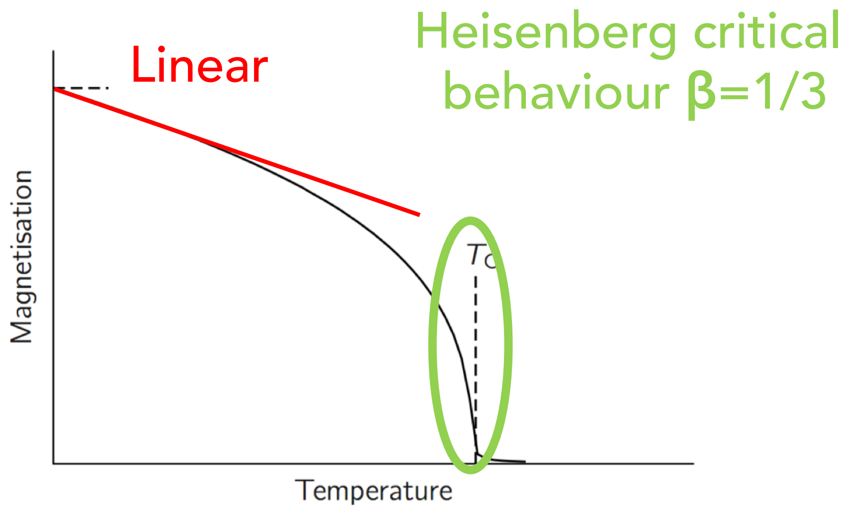
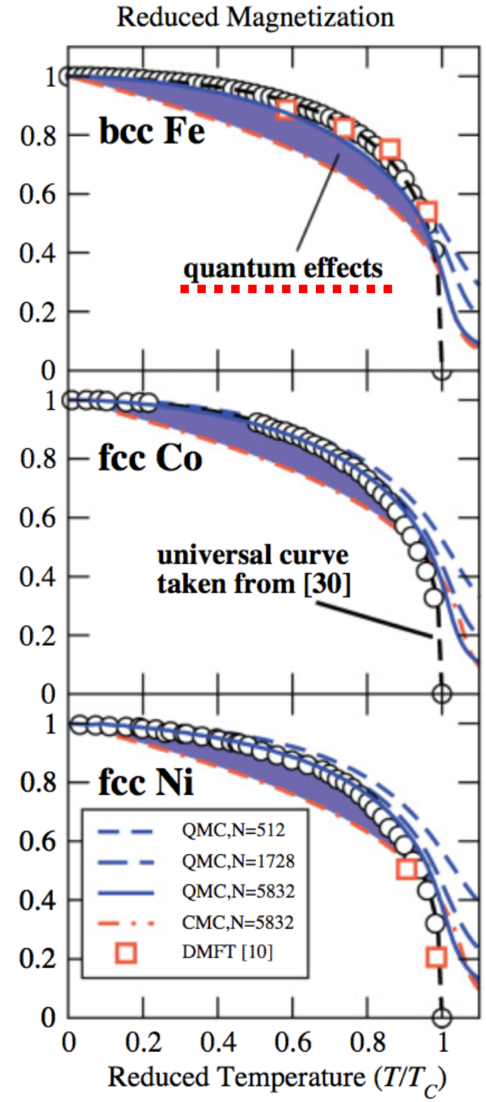
Experiments



Simulations (ASD or Monte Carlo)





Körmann et al. *Phys. Rev. B* **83,** 165114 (2011)

Magnon specific heat capacity

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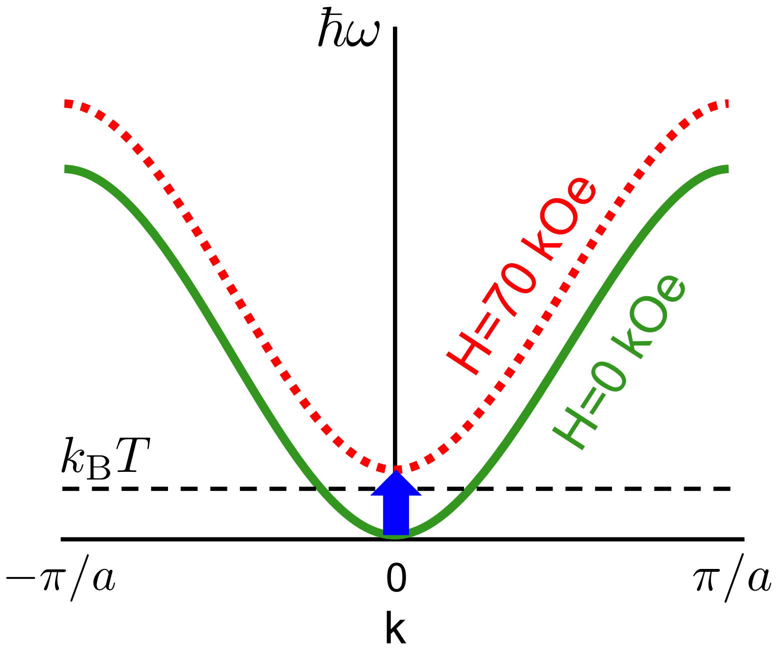
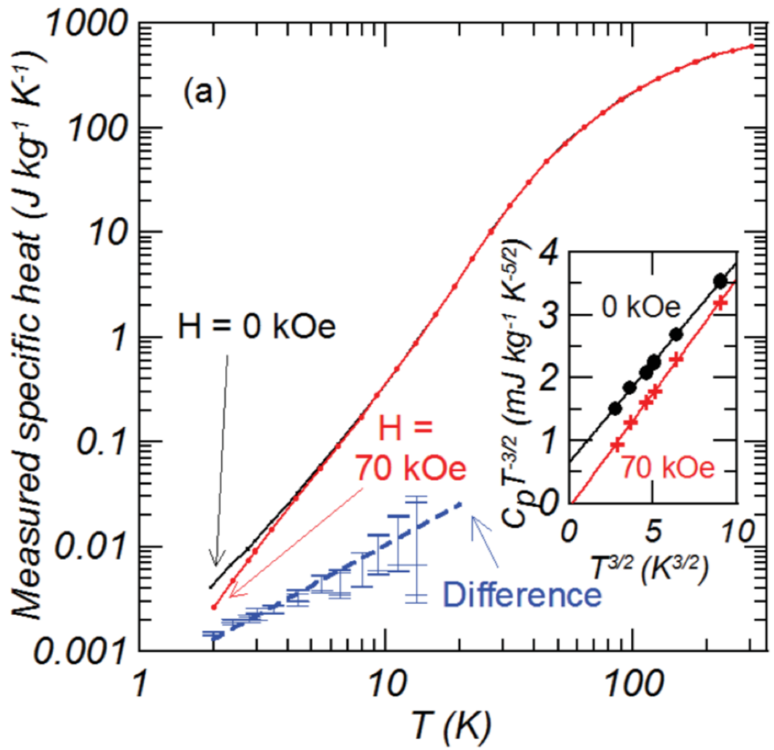
Only measurable at low temperatures

Measurement only possible where the magnons can be frozen by an external field

$$\kappa_m = \frac{1}{3} C_m v_m l_m$$





Boona *et al. Phys. Rev. B* **90**, 064421 (2014)

Quantum thermodynamics through T rescaling

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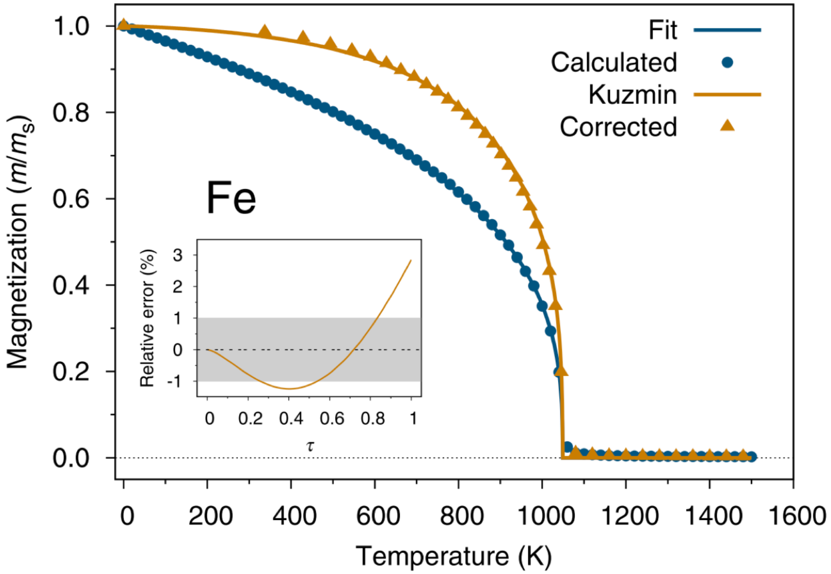
Evans et al. *Phys. Rev. B* **91,** 144425 (2015)

Woo et al. *Phys. Rev. B* **91,** 104306 (2015)

$$\frac{T_{sim}}{T_c} = \left(\frac{T_{exp}}{T_c}\right)^{\alpha}$$



- The opposite of ab initio
- Does not work for most thermodynamic quantities

$$\eta_S(T) = \int_0^\infty \frac{\hbar\omega}{\exp(\hbar\omega/k_B T) - 1} g_m(\omega, T) d\omega$$

$$g_m(\omega, T) = \frac{\Omega}{(2\pi)^3} \frac{4\pi k^2}{\nabla_k \omega(T)}$$

- Requires good approximation of spin wave spectrum
- May still have issues with magnon populations and lifetimes

Quantum statistics in ASD

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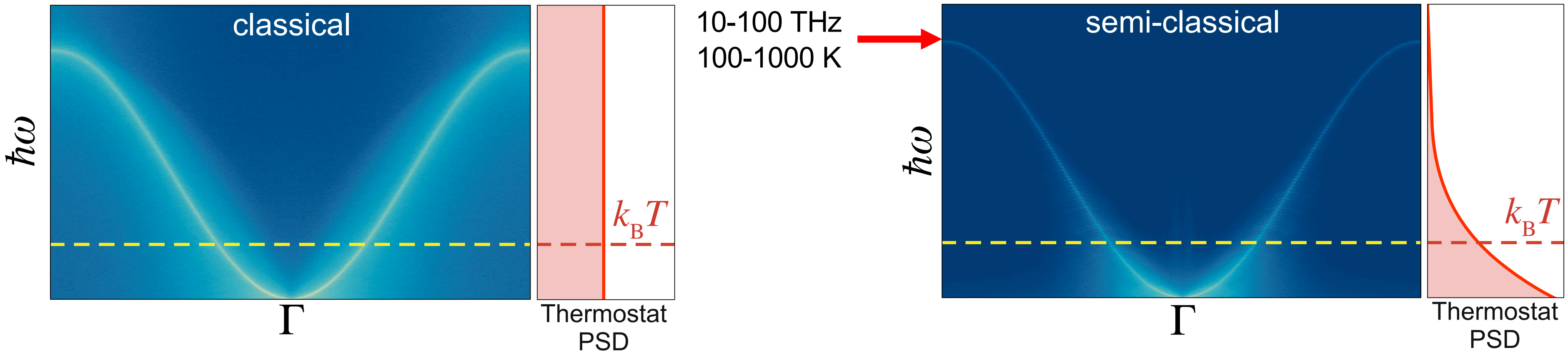
Atomistic spin dynamics does not work well at “low temperatures”.



$$\Theta(\omega) \propto k_B T$$

$$\Theta(\omega) \propto \frac{\hbar\omega / k_{\mathrm{B}} T}{e^{\hbar\omega / k_{\mathrm{B}} T} - 1}$$

$\hbar|\omega| \ll k_B T$ Classical limit is only true for part of the spectrum

Overpopulation of high frequency / high wavenumbers

General Langevin equation

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$$\mathbf{H}_i(t) = -\frac{\partial \mathcal{H}}{\partial \mathbf{S}_i} - \eta \int_{-\infty}^{t} \varphi(t-t')\frac{d\mathbf{S}_i}{dt'}dt' + \boldsymbol{\xi}_i(t)$$

Potential — Friction — Noise

$$\varphi(|t-t'|) = \delta(|t-t'|)$$

Markovian process - damping is local in time

$$\langle \xi_{i,a}(t) \rangle = 0$$

$$\langle \xi_{i,a}(t)\xi_{j,b}(t') \rangle = 2\eta\Theta(|t-t'|)$$

Classical limit – fluctuation dissipation theorem

$$\Theta(\omega) \propto k_B T$$

White noise – independent of frequency

Quantum mechanics

$$\Theta(\omega) \propto \frac{\hbar\omega}{2} + \frac{\hbar\omega}{\exp(\hbar\omega/k_B T) - 1}$$

Colored noise – frequency dependent

Semi-Quantum Spin Dynamics

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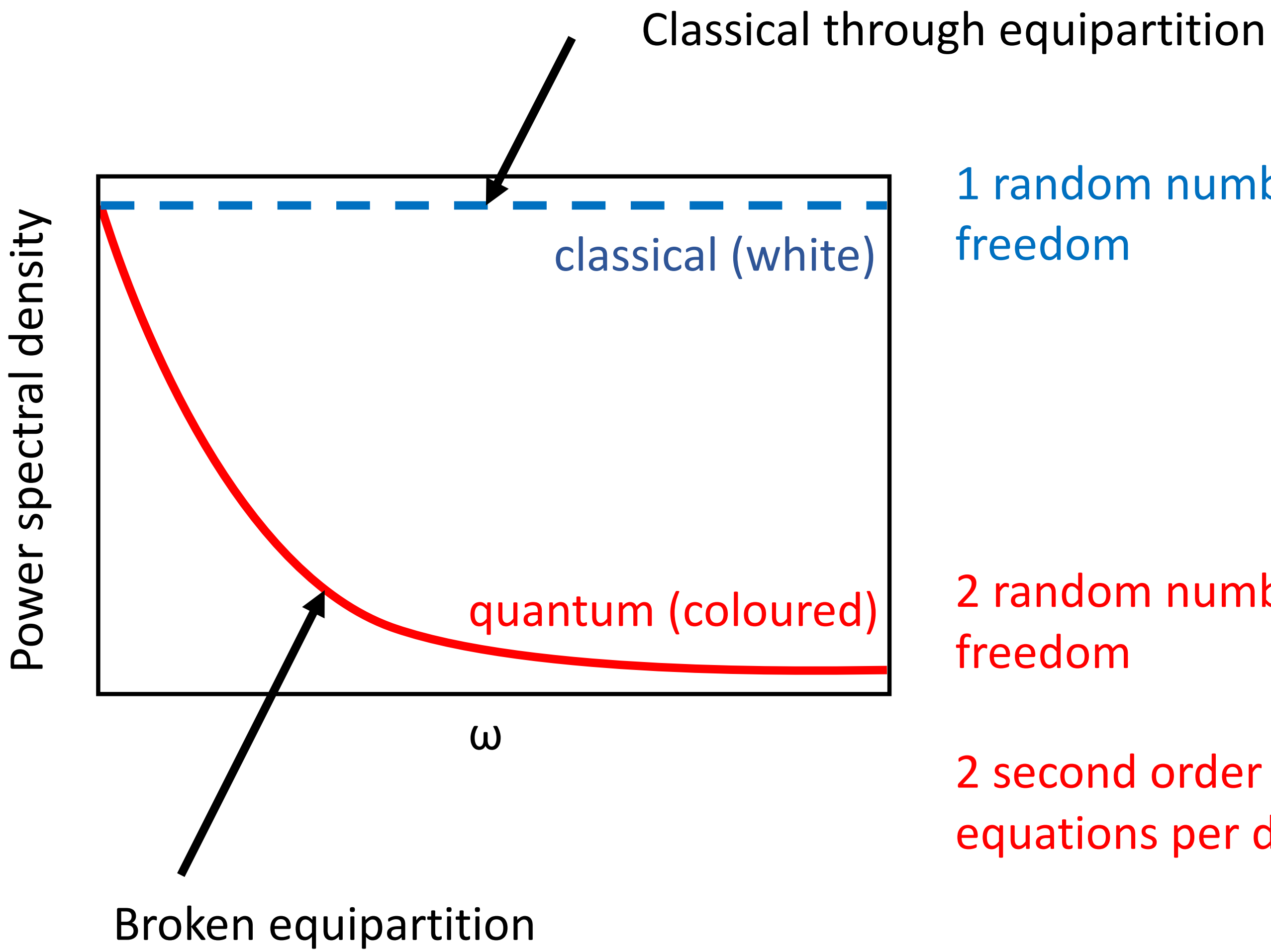
Approximate coloured spectrum with multiple stochastic differential equations

Savin et al. Phys. Rev. B **86**, 064305 (2012)



1 random number per degree of freedom

2 random numbers per degree of freedom

2 second order differential equations per degree of freedom

Barker et al. arXiv:1902.00449 (2019)

Validation for a simple ferromagnet

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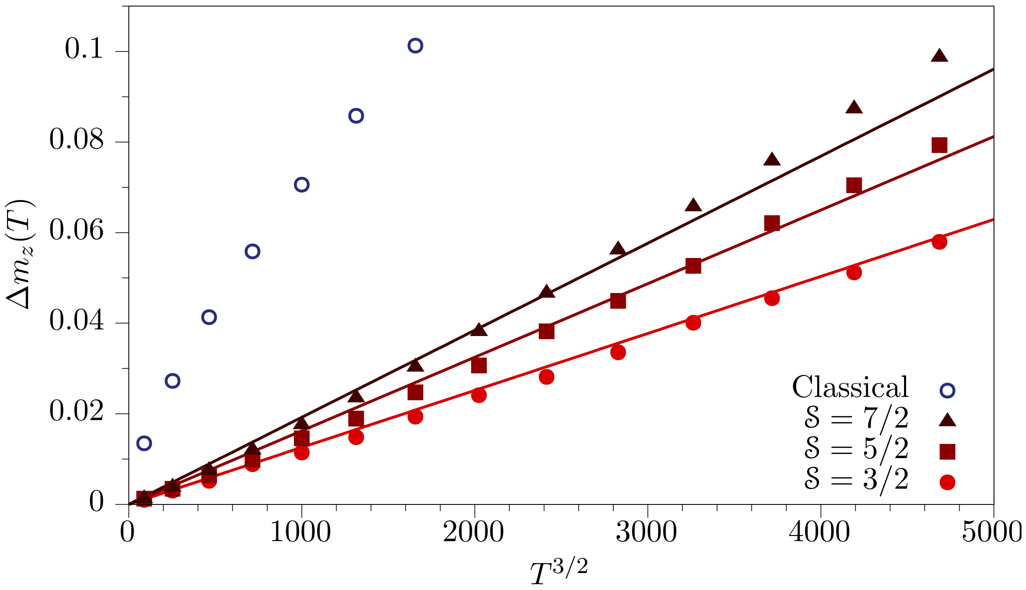
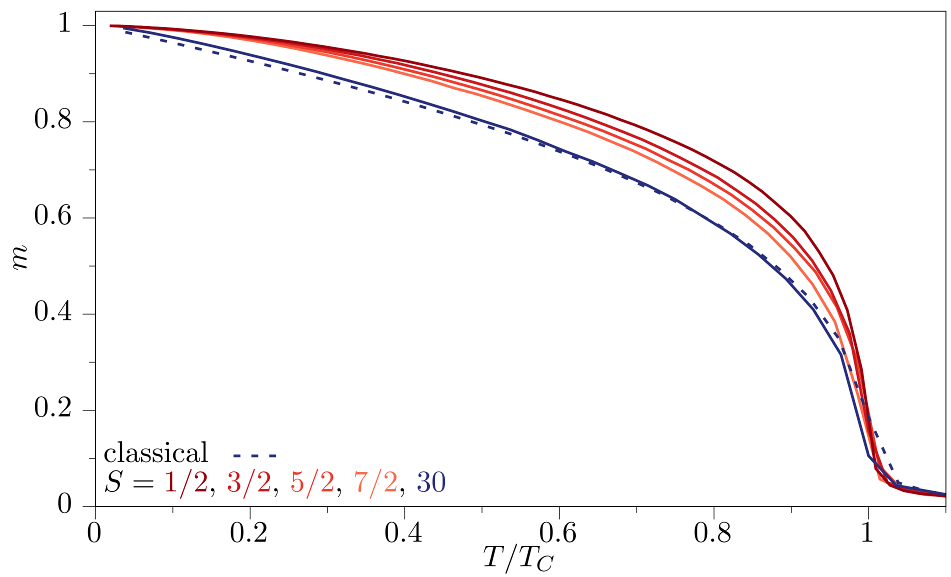
Low T magnetization - Bloch's $T^{3/2}$ law



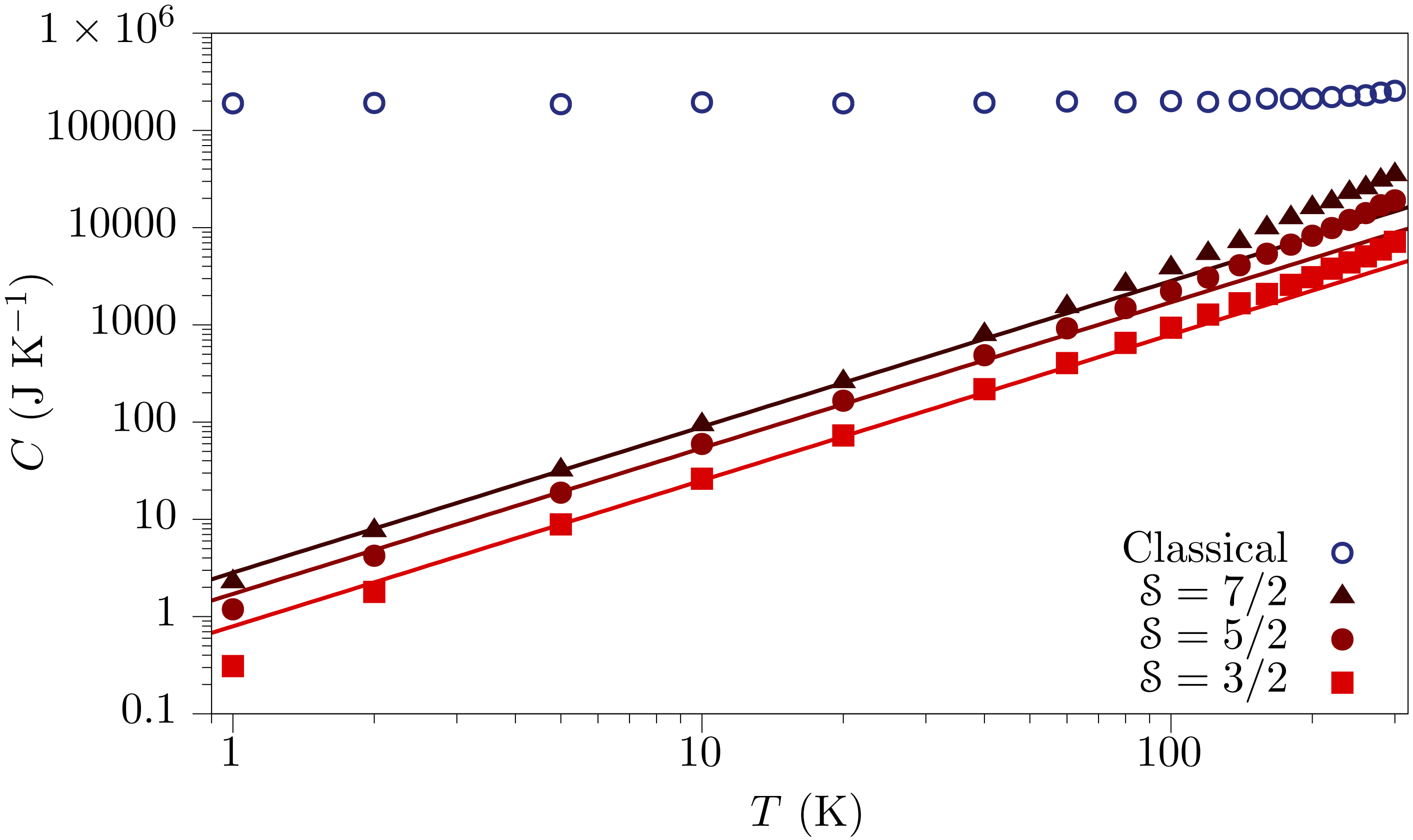
$$\Delta m_z(T) = 1 - m_z(T) = \frac{1}{\mathcal{S}} \sum_{\mathbf{k}} \langle n_{\mathbf{k}} \rangle$$

$$\Delta m_z(T) = v_{\mathrm{ws}} \frac{1}{\mathcal{S}} \frac{\Gamma\left(\frac{3}{2}\right)\zeta\left(\frac{3}{2}\right)}{2\pi^2} \left(\frac{k_B T}{D}\right)^{3/2}$$

Shape of magnetization



Magnon heat capacity



$$C(T) = \frac{1}{V}\frac{\partial}{\partial T}\sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}} \langle n_{\mathbf{k}} \rangle$$

$$C(T) = \frac{5}{8}\frac{\Gamma\left(\frac{5}{2}\right)\zeta\left(\frac{5}{2}\right)}{\pi^2} k_B \left(\frac{k_B T}{D}\right)^{3/2}$$

S-dependence of Curie temperature

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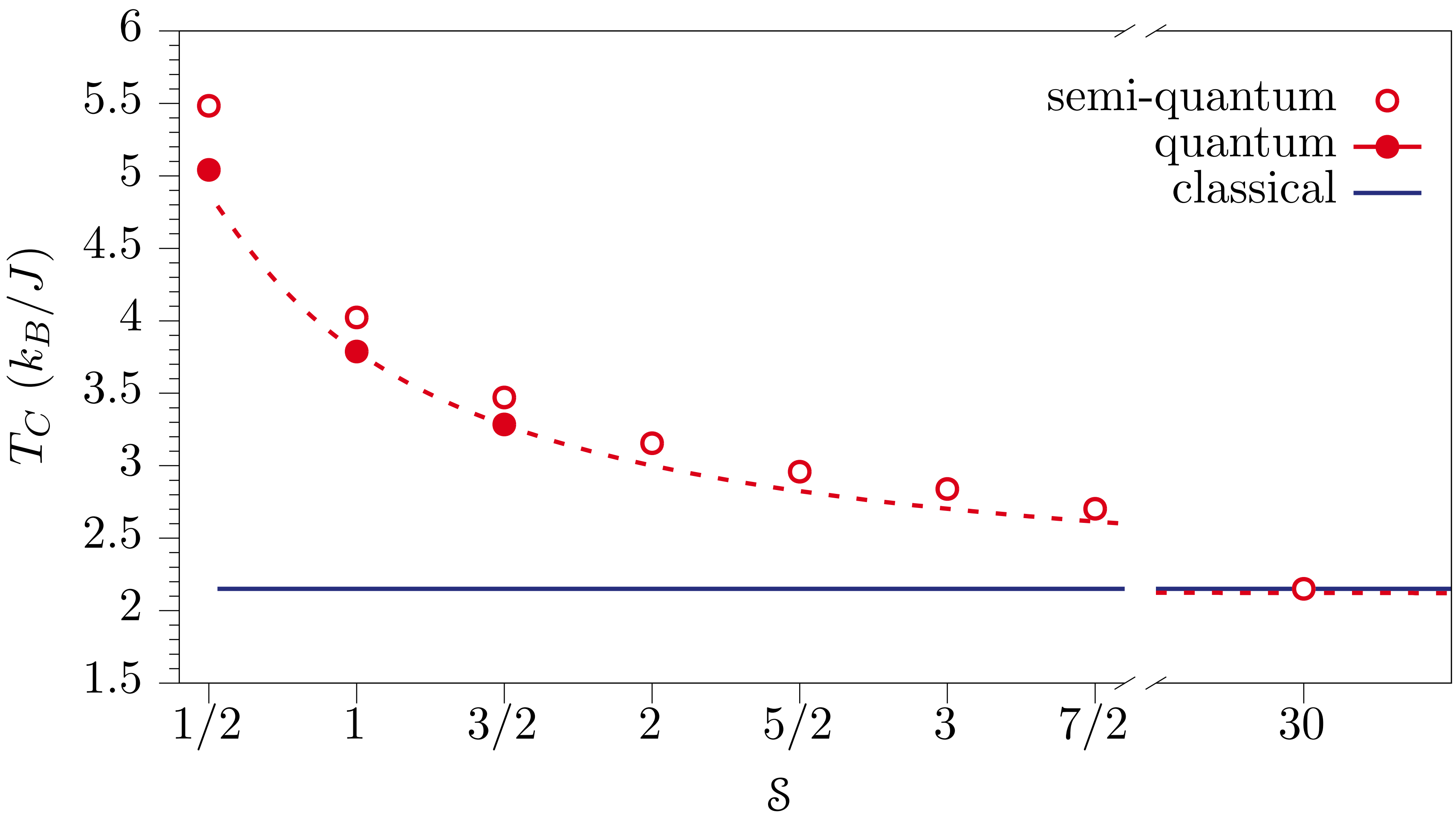
In classical systems T_c depends only on J_{ij}

In quantum systems T_c depends also on S



Quantum from high temperature series expansion - Oitmaa, et al. *J. Phys. Condens. Matter* **16,** 8653 (2004)

Be very careful with factors of 2 (and the sign)

$$\mathcal{H} = -\sum_{i<j} \mathbf{S}_i \mathbb{J}_{ij} \mathbf{S}_j \qquad \mathcal{H} = -2\sum_{i<j} \mathbf{S}_i \mathbb{J}_{ij} \mathbf{S}_j$$

$$\mathcal{H} = -\frac{1}{2}\sum_{i\neq j} \mathbf{S}_i \mathbb{J}_{ij} \mathbf{S}_j \qquad \mathcal{H} = -\sum_{i\neq j} \mathbf{S}_i \mathbb{J}_{ij} \mathbf{S}_j$$

Know which Hamiltonian you are using for input!

Yttrium Iron Garnet

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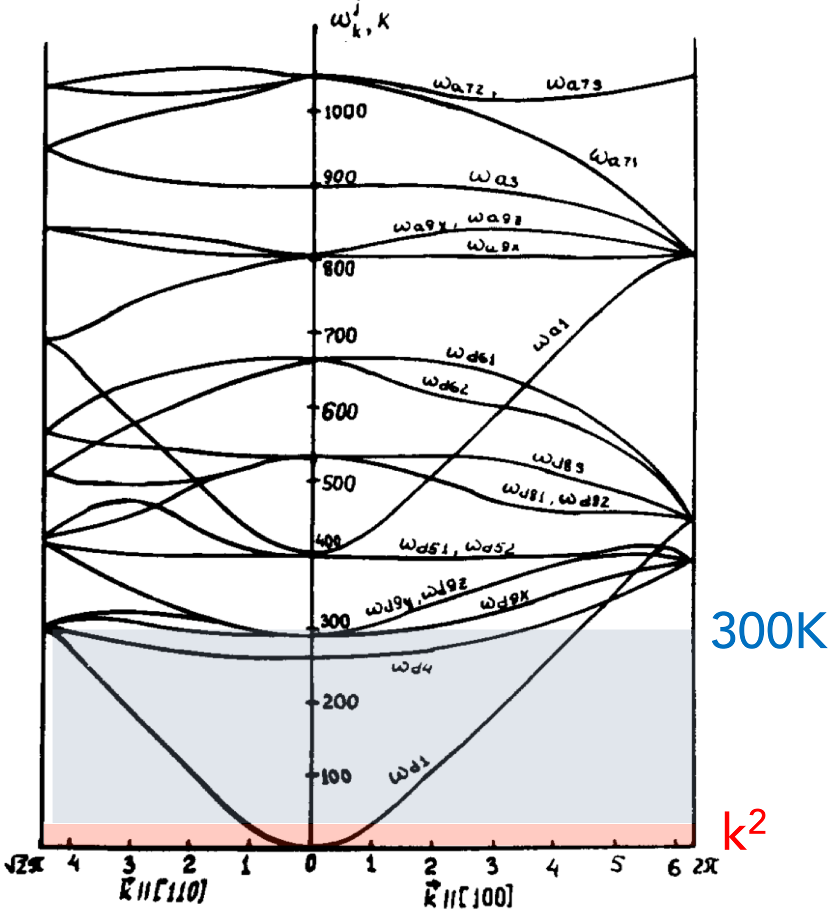
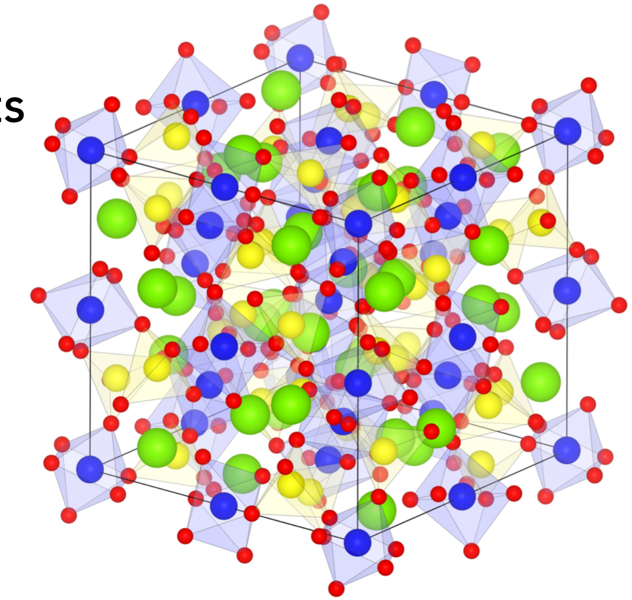
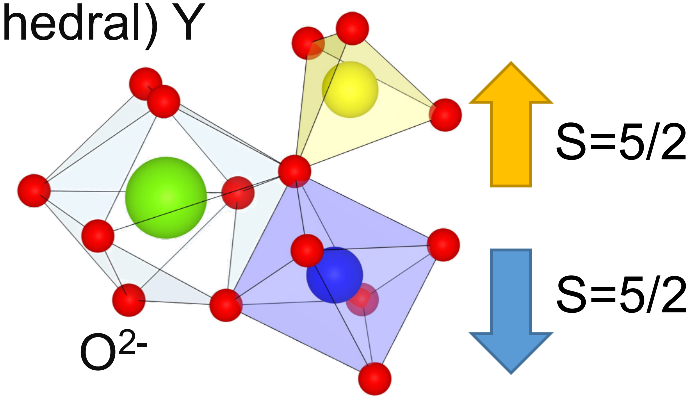
Very large unit cell

20 Fe atoms in primitive cell in two different environments



Cherepanov et al. *Phys. Rep.* **229,** 81 (1993)





Gilbert damping
$\alpha = 0.00001$

New YIG Exchange Parameters

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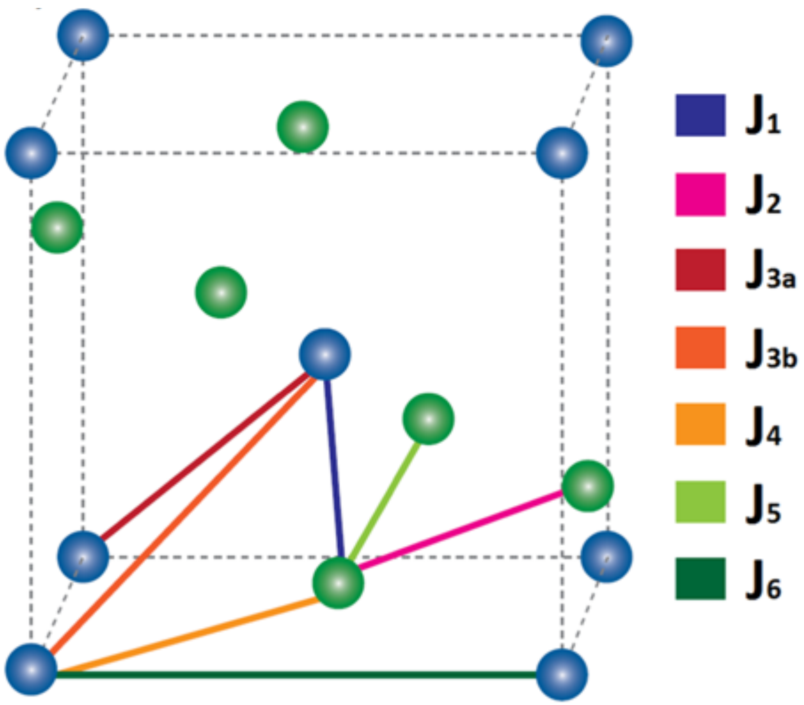
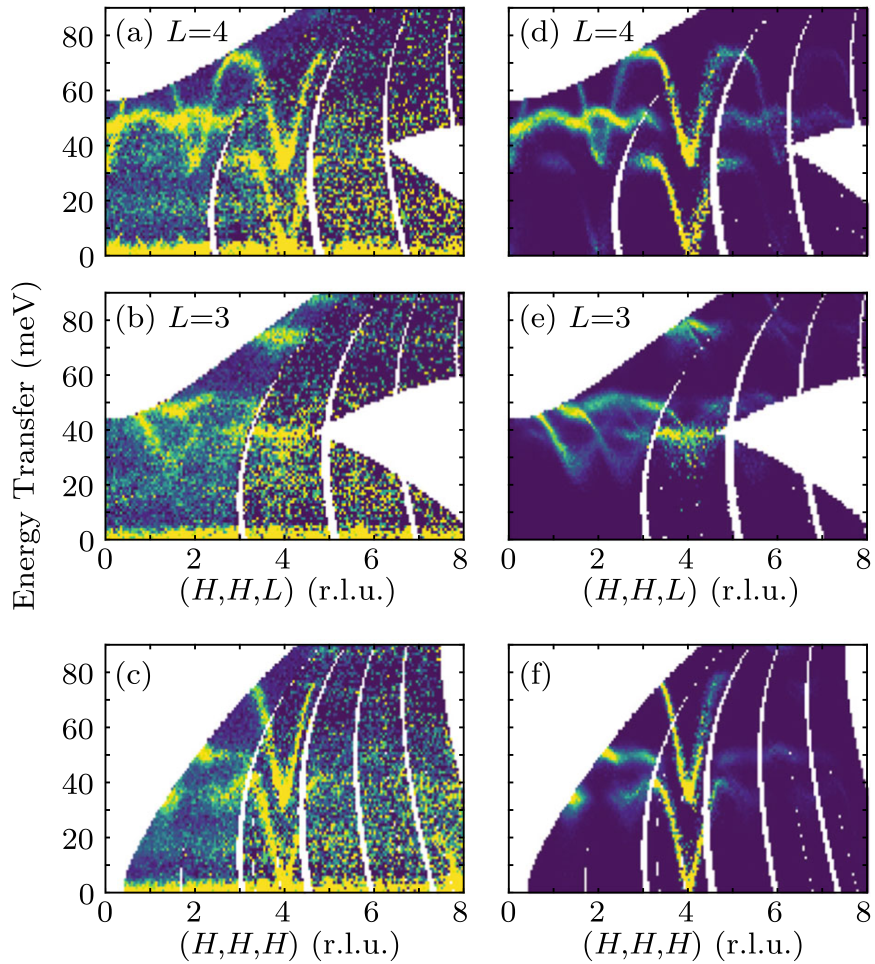


Exchange	Energy (meV)
J_1	6.8(2)
J_2	0.52(4)
J_{3a}	0.0(1)
J_{3b}	1.1(3)
J_4	–0.07(2)
J_5	0.47(8)
J_6	–0.09(5)



Princep *et al. npj Quantum Mater.* **2,** 63 (2017)

Temperature dependent magnetization

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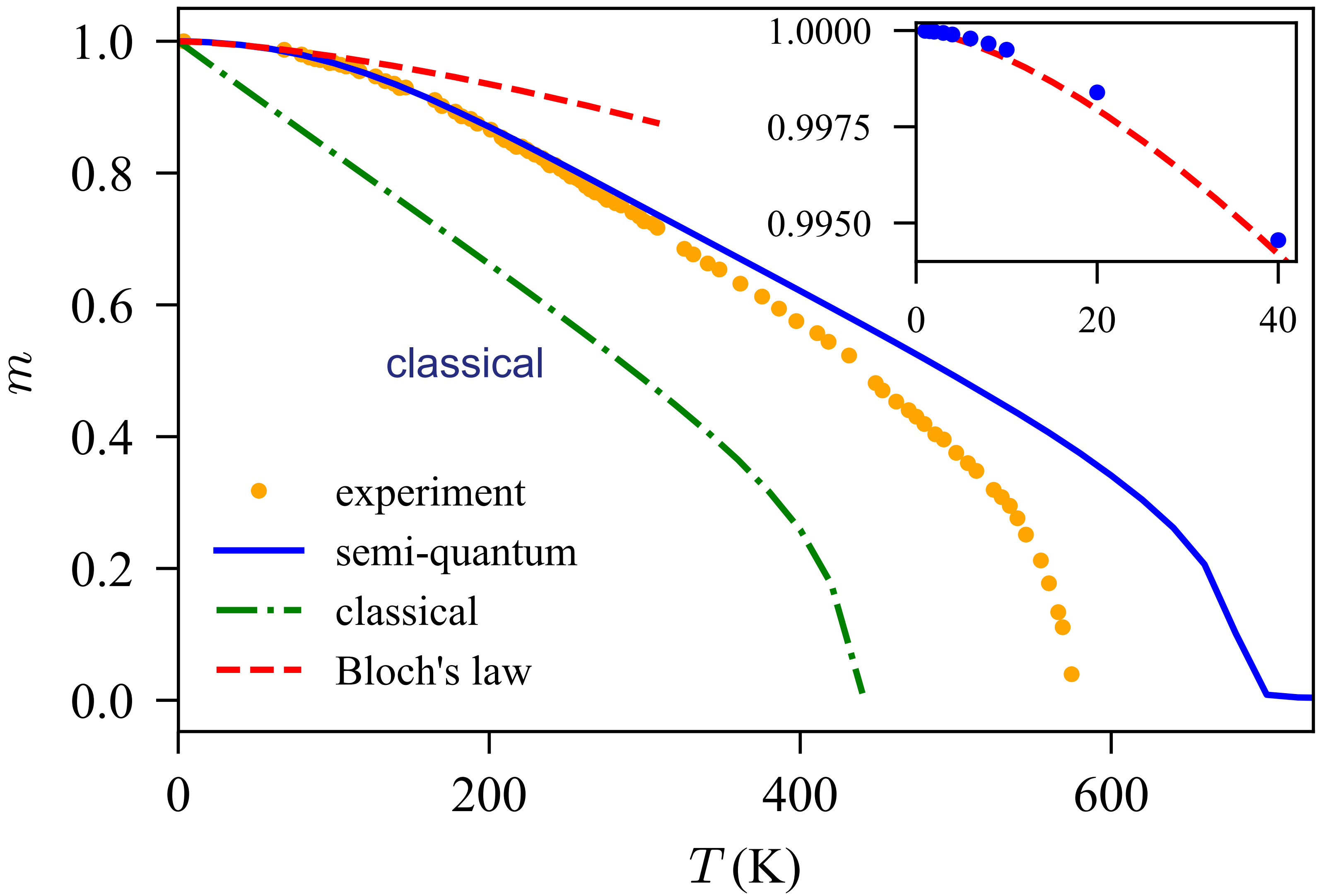

Experiment
Anderson, *Phys. Rev.* **134**, A1581 (1964)

Barker et al. arXiv:1902.00449 (2019)

What happens to the spectrum with quantum noise?

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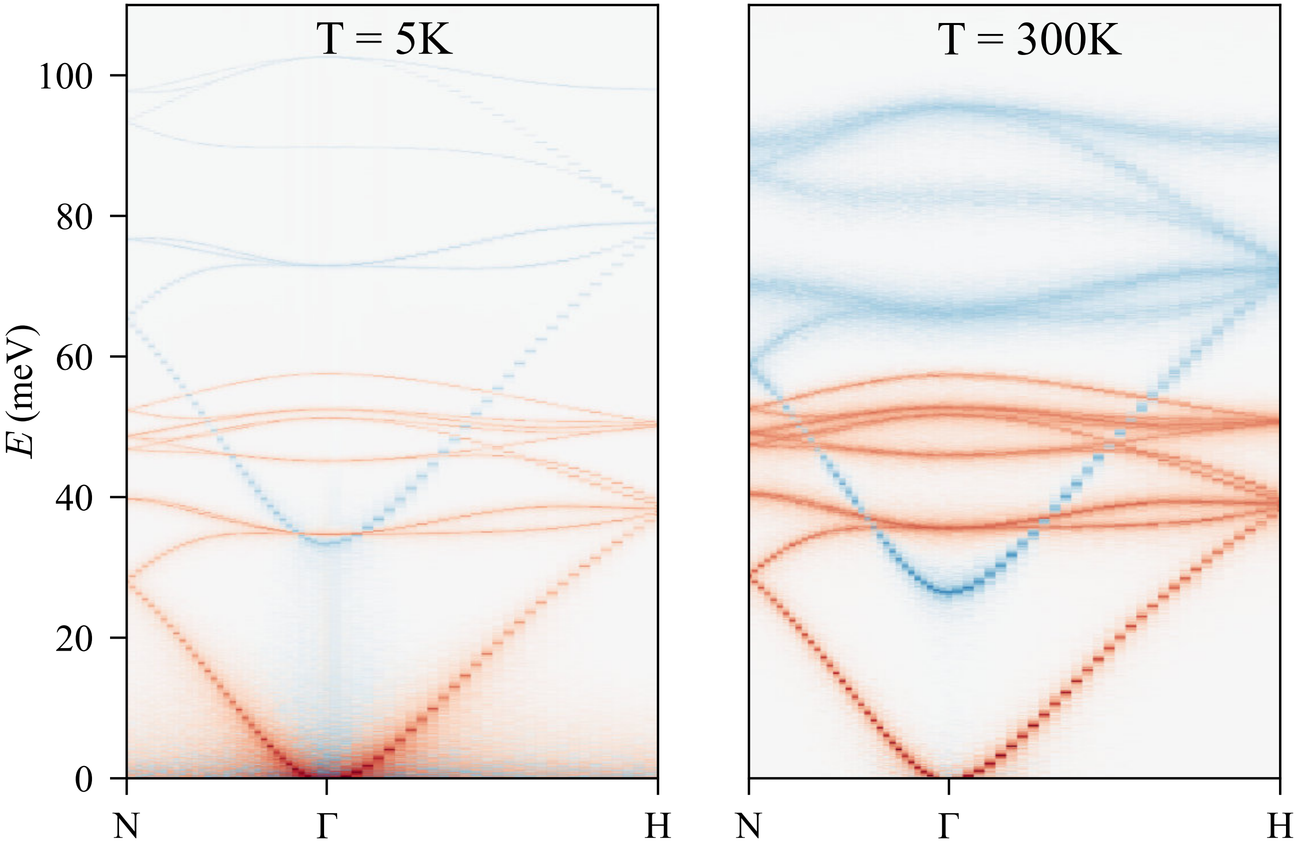
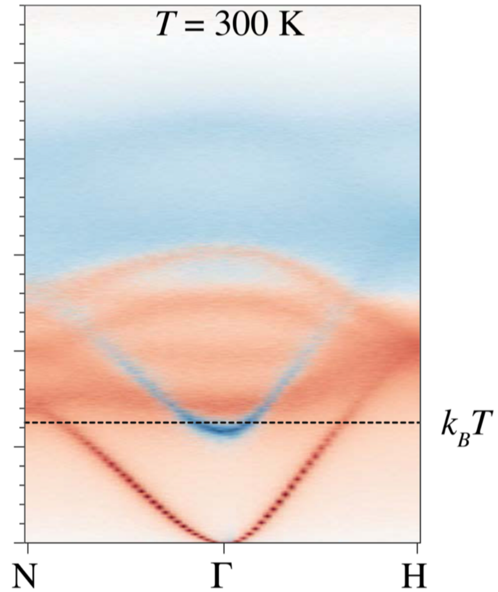


Classical result



The role of magnon polarization in the spin Seebeck effect

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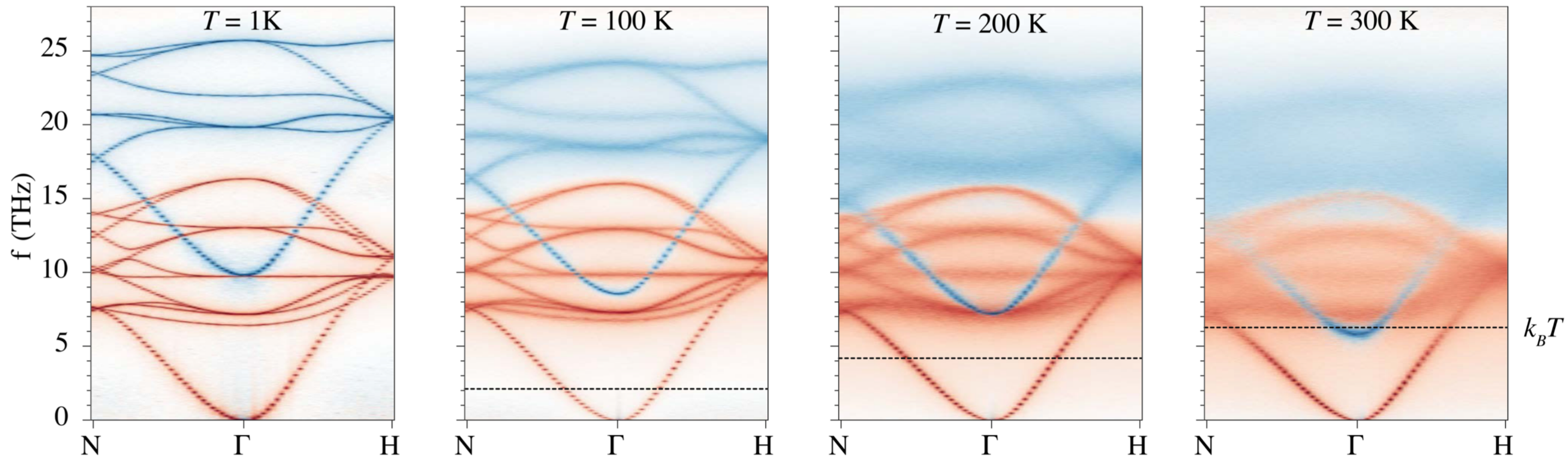
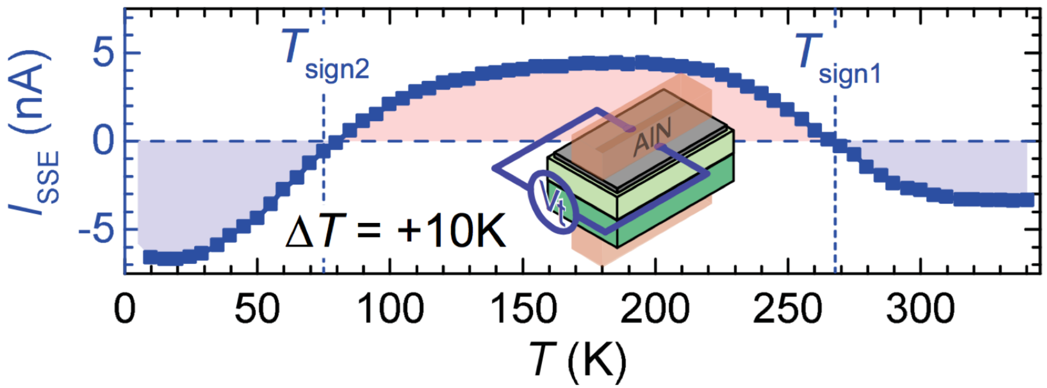
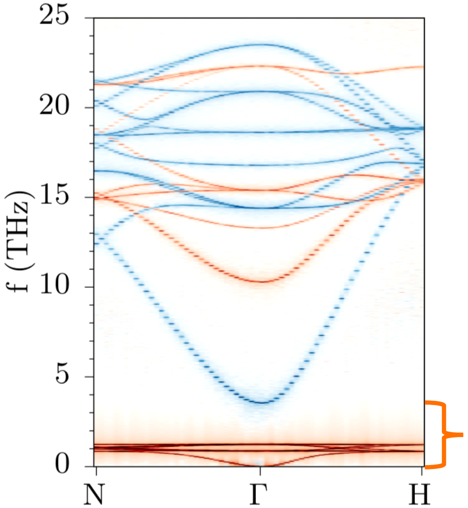
$$\mathcal{S}_{\alpha\beta}(\mathbf{q},\omega) = \frac{1}{N\sqrt{2\pi}} \sum_{\mathbf{r},\mathbf{r}'} \mathrm{e}^{\mathrm{i}\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}')} \int_{-\infty}^{+\infty} \mathrm{e}^{\mathrm{i}\omega t} \langle S_\alpha(\mathbf{r},t) S_\beta(\mathbf{r}',0)\rangle dt$$



Barker *et al. Phys. Rev. Lett.* **117**, 217201 (2016)





Geprägs. *et al. Nat. Commun.* **7,** 10452 (2016)

Magnon specific heat capacity

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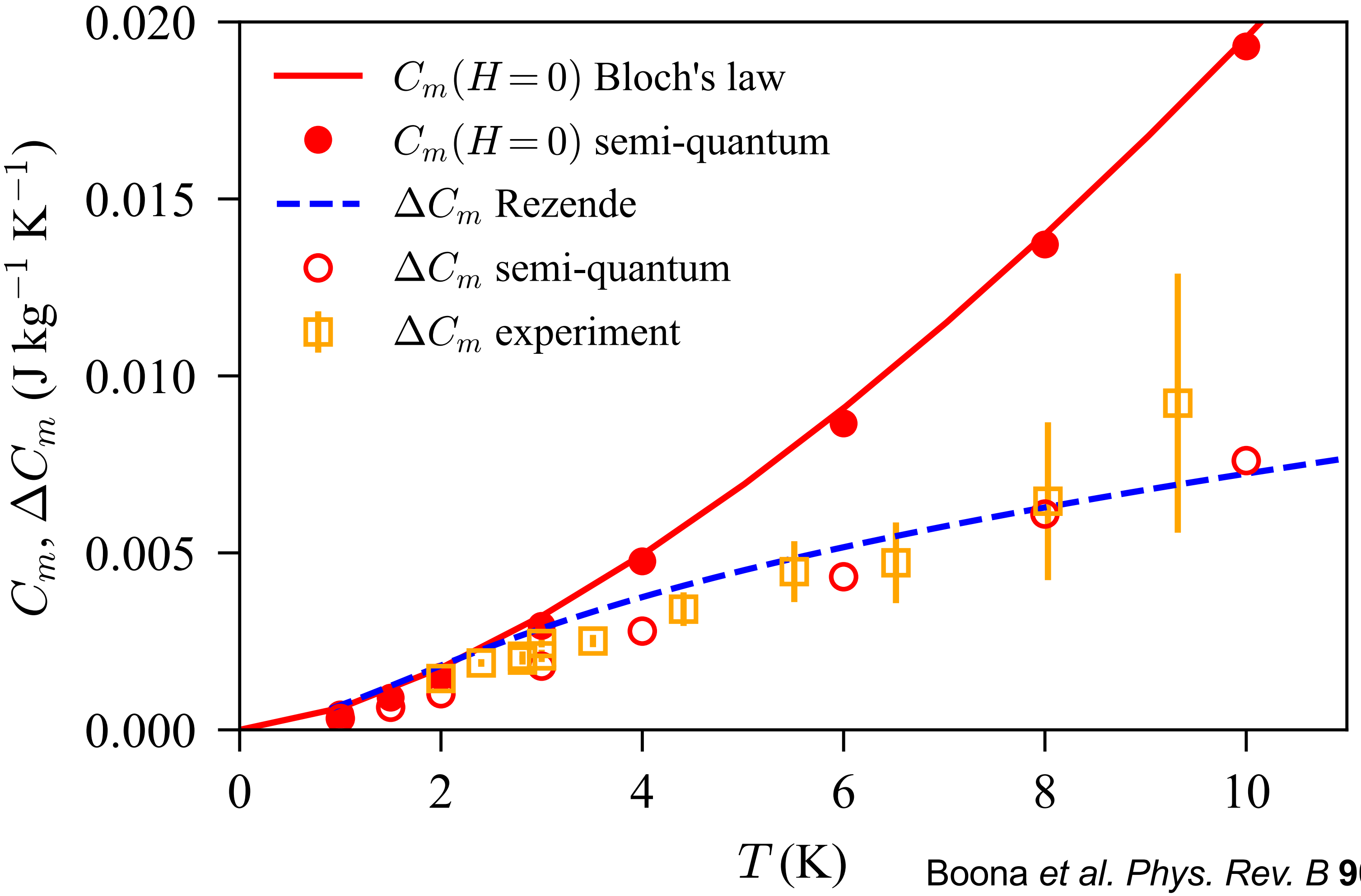
Quantitative agreement

Simulations and experiments are entirely independent of one another



Boona *et al. Phys. Rev. B* **90**, 064421 (2014)
Rezende *et al. Phys Rev B*, ***91***, 104416 (2015)
Barker et al. arXiv:1902.00449 (2019)

Magnon specific heat capacity

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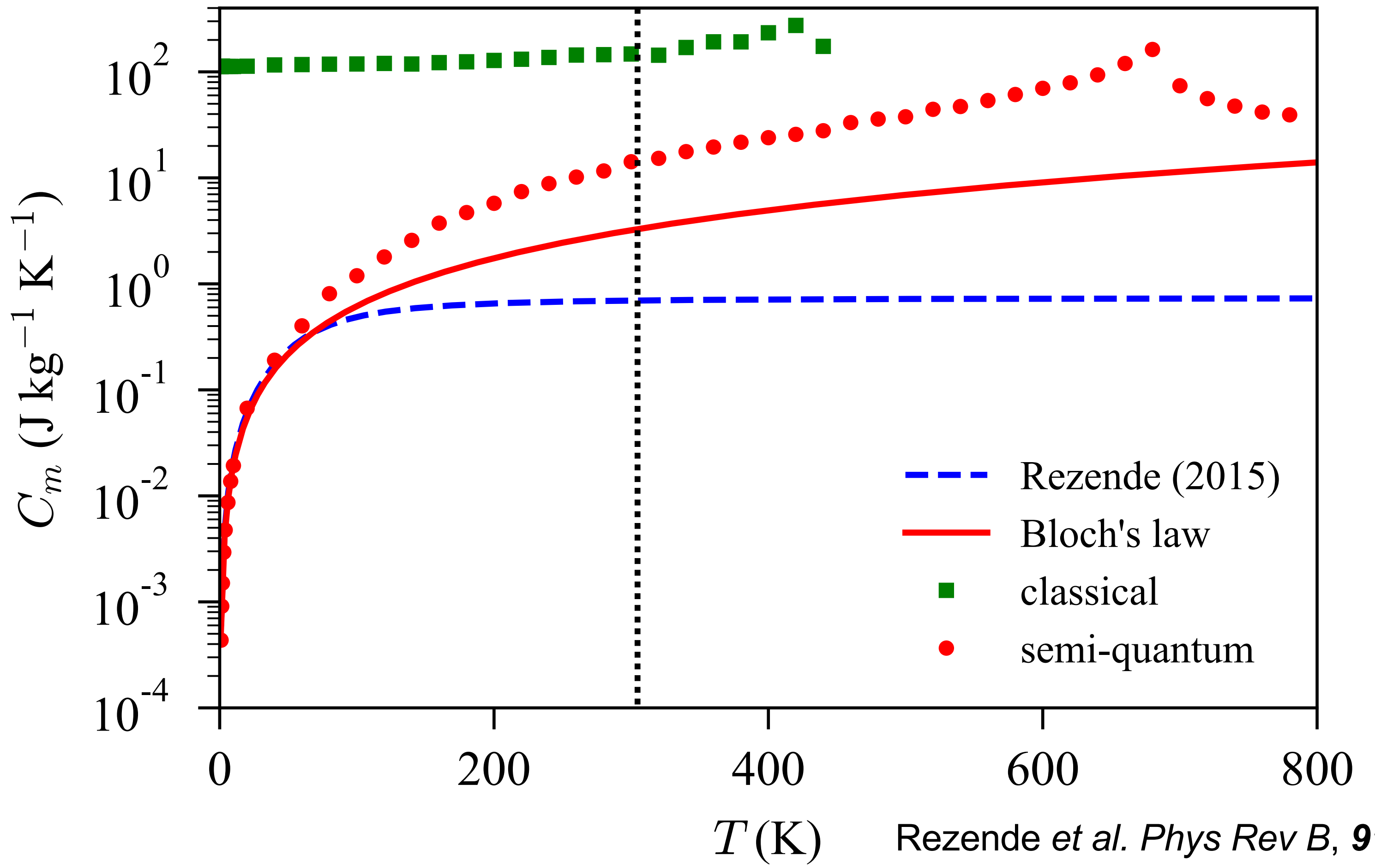
Specific heat at higher temperatures

The assumption of a ferromagnetic k^2 dispersion is wrong

Ignoring the optical modes is also wrong



Rezende *et al. Phys Rev B*, ***91***, 104416 (2015)

Barker et al. arXiv:1902.00449 (2019)

Non-local spin transport

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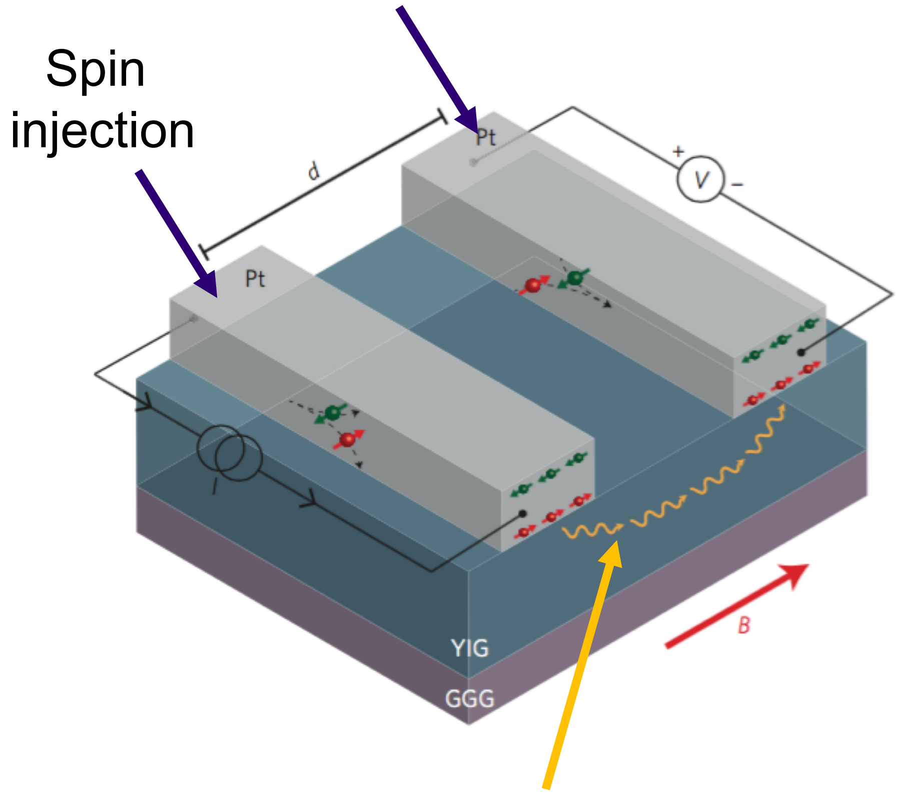
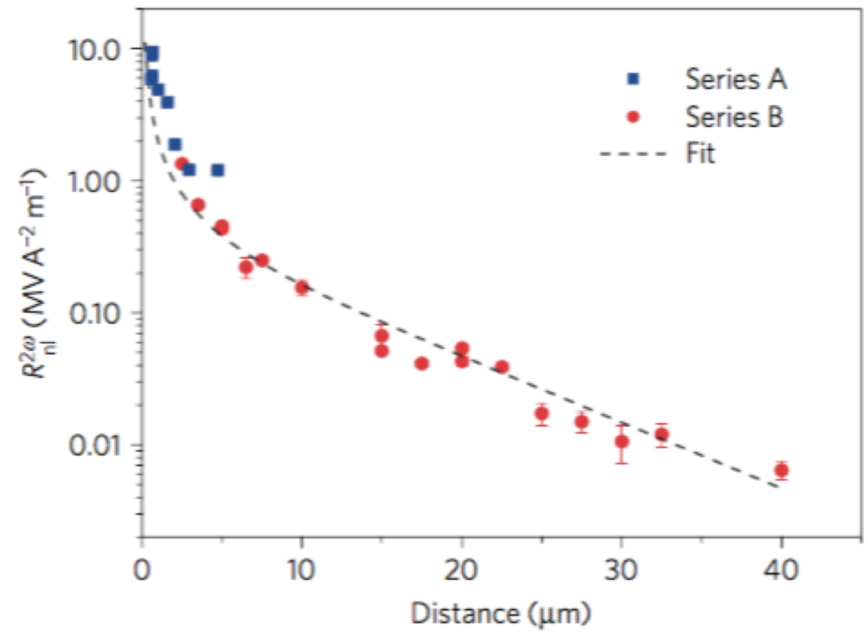
Long range magnon spin transport

Non local signal of spin transport in YIG over microns





Cornelissen *et al. Nat. Phys.* **11**, 1022 (2015)

Magnon transport properties

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Boltzmann theory of magnon spin transport

Theory makes many assumptions about timescales and transport properties

Onsager Matrix

$$\begin{pmatrix} \frac{2e}{\hbar}\mathbf{j}_m \\ \mathbf{j}_{\mathrm{Q,m}} \end{pmatrix} = -\begin{pmatrix} \sigma_m & L/T \\ \hbar L/2e & \kappa_m \end{pmatrix} \begin{pmatrix} \nabla \mu_m \\ \nabla T_m \end{pmatrix}$$

magnon spin conductivity
magnon heat conductivity
bulk spin Seebeck coefficient

Cornelissen *et al. Phys. Rev. B* **94**, 014412 (2016)

Problem: the transport coefficients are very difficult to measure in an experiment

Magnon spin conductivity

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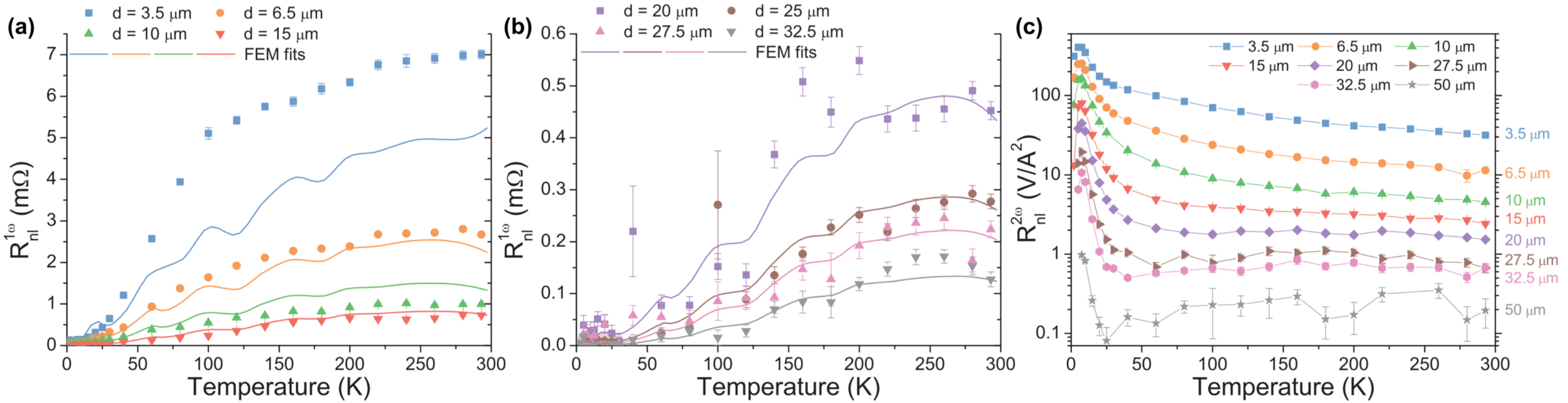
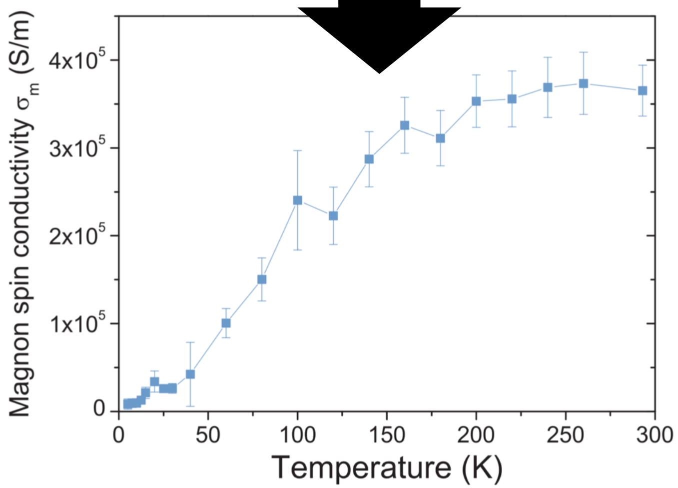
Very difficult to extract from experiments



Boltzmann assumptions + COMSOL + fitting



Cornelissen *et al. Phys. Rev. B* **94**, 180402(R) (2016)

Kubo formula

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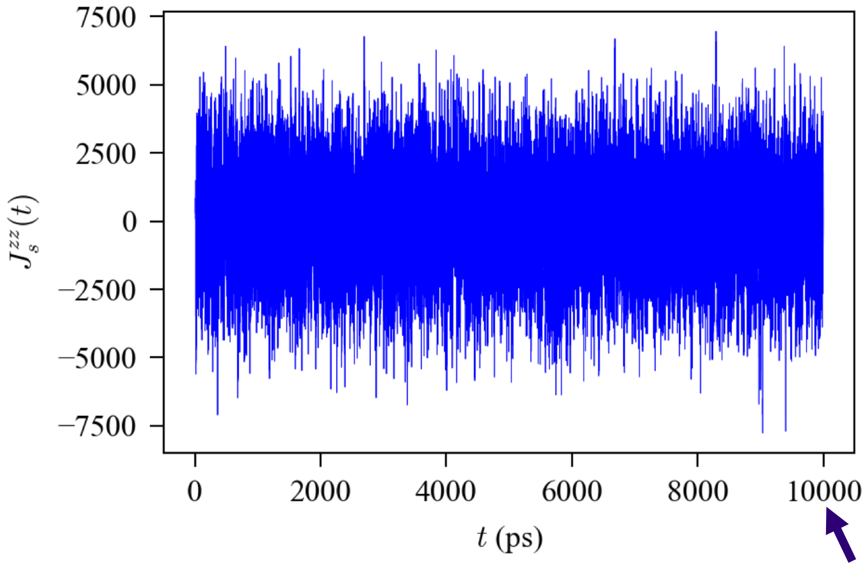
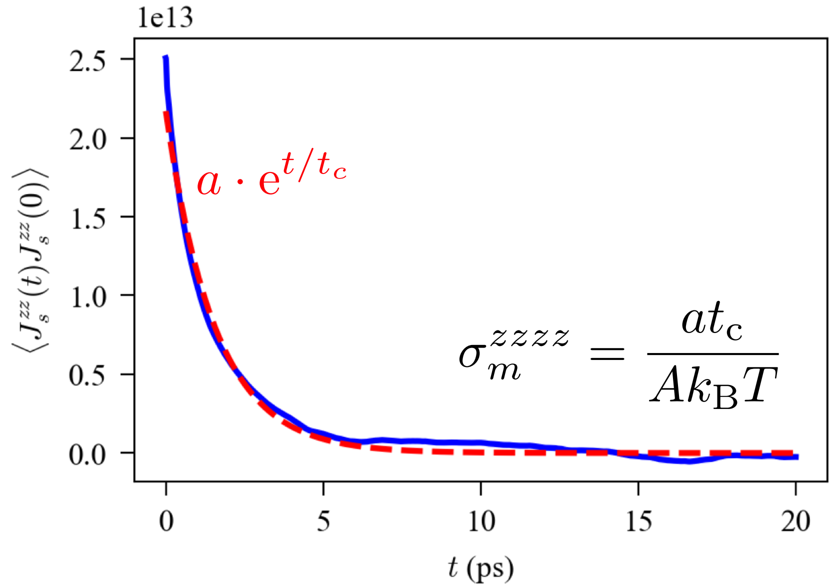
Conductivity can be calculated from current correlations

$$\sigma_m^{\alpha\beta\mu\nu} = \int_0^\infty \frac{1}{Ak_\mathrm{B}T}\langle J_s^{\alpha\beta}(t)J_s^{\mu\nu}(0)\rangle dt$$

μ-component of spin current in the ν-direction

$$J_s^{zz} = \gamma \sum_{i<j} J_{ij}\left(\hat{z}\cdot\mathbf{r}_{ij}\right)\left(\hat{z}\cdot\left(\mathbf{S}_i \times \mathbf{S}_j\right)\right)$$





100,000,000 timesteps

Requires very long integration times

Magnon spin conductivity

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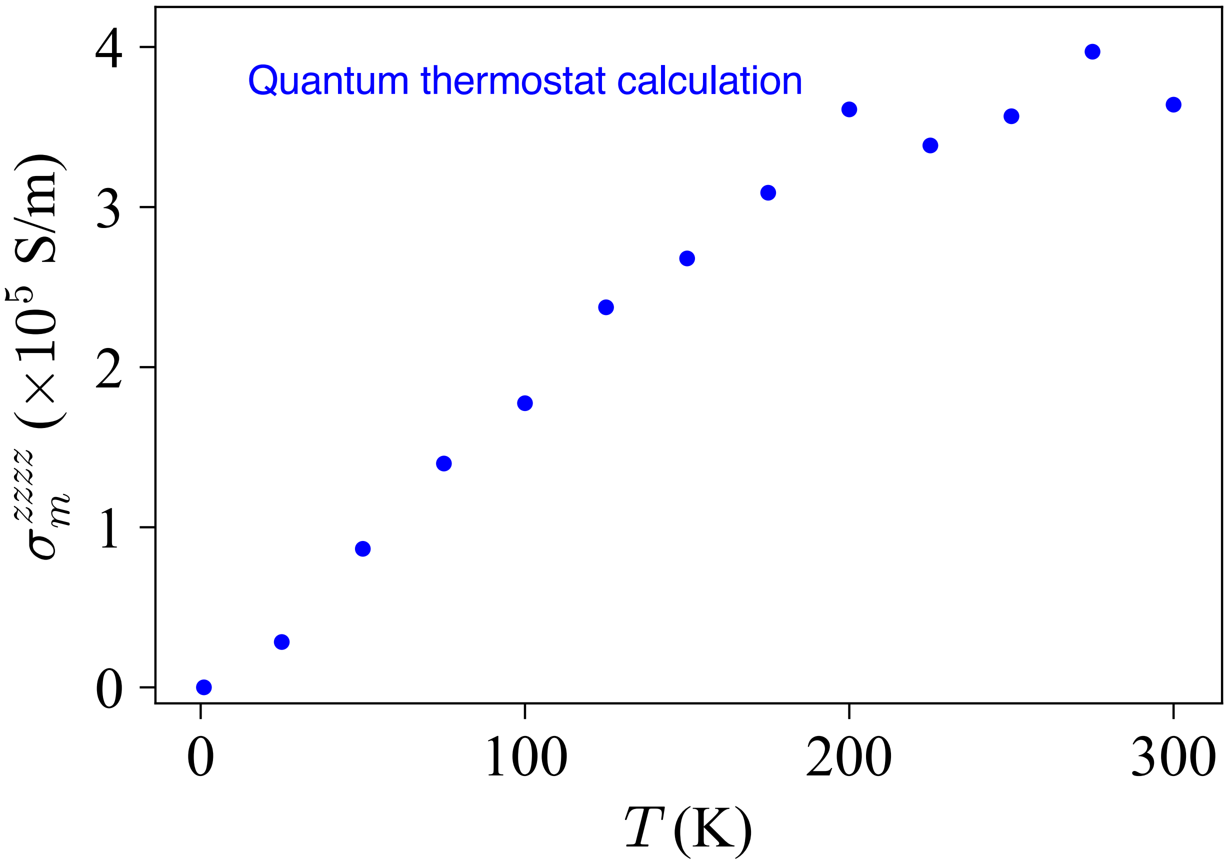
Temperature dependence from simulation



Magnon spin conductivity

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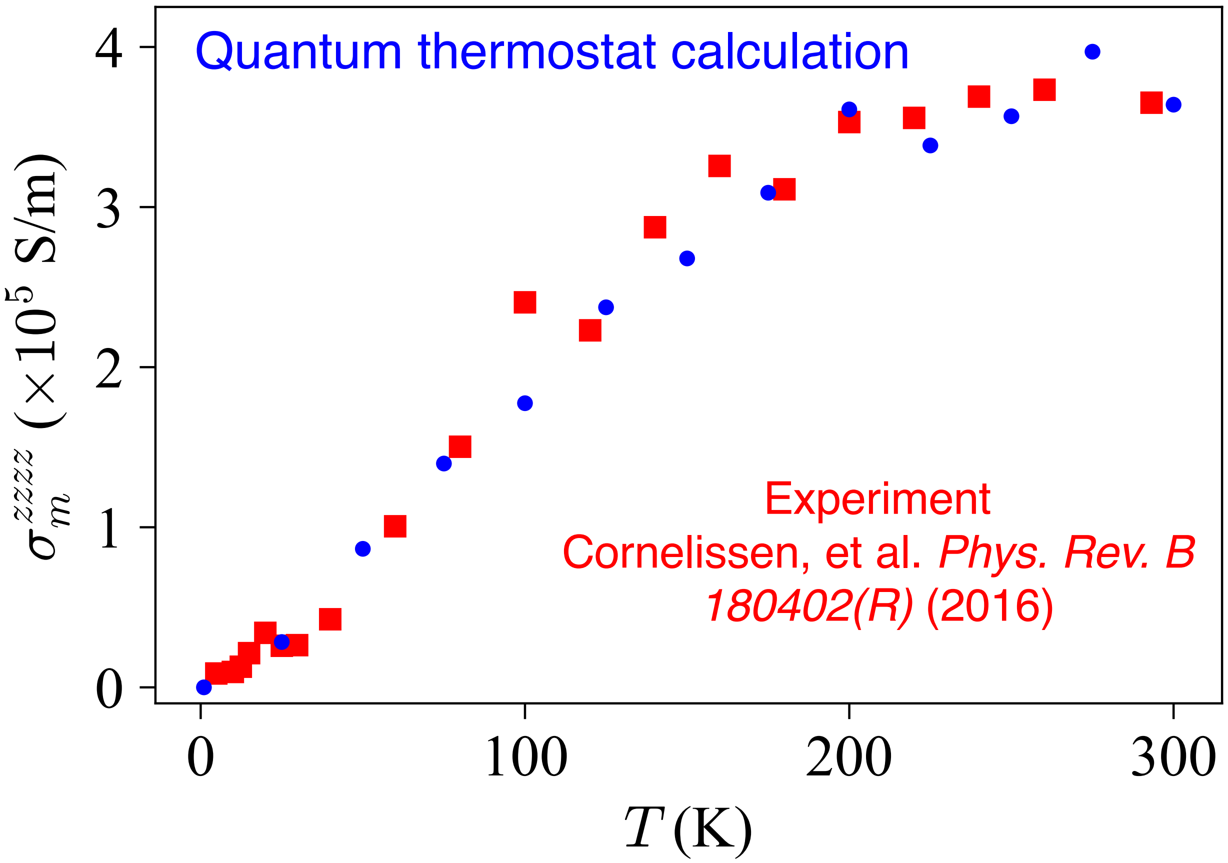
Excellent quantitative agreement



Acknowledgements

Collaborators

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- Takashi Kikkawa

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