20th Mardi Gras Conference

Towards a numerically exact description of correlated open quantum systems

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Presenting the results obtained in cooperation with
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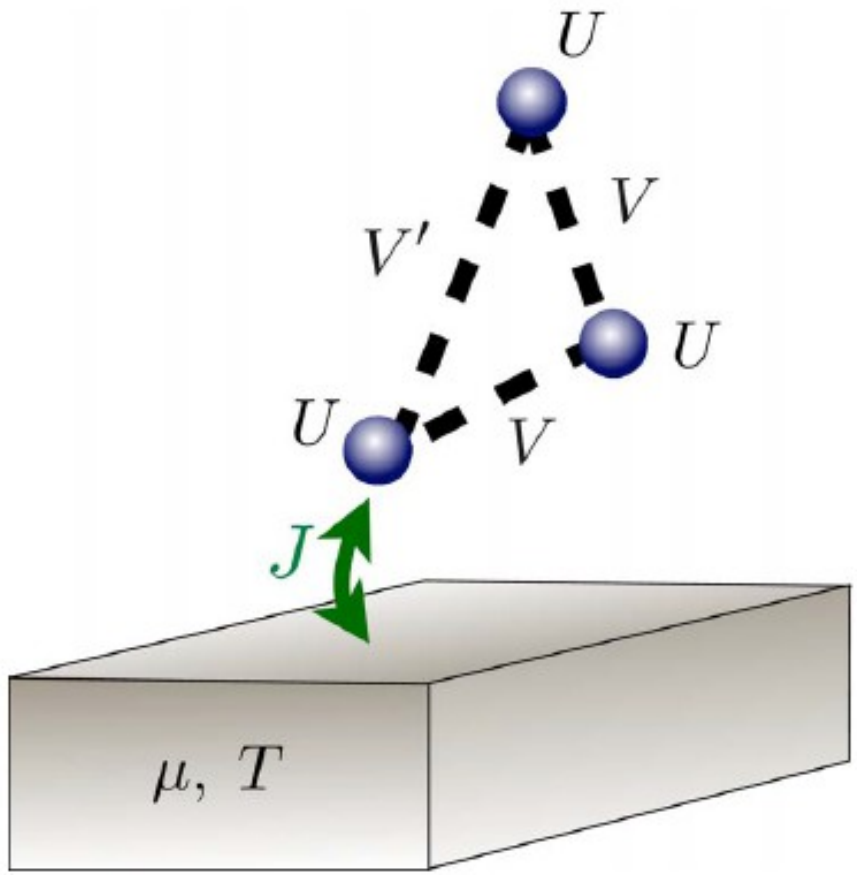
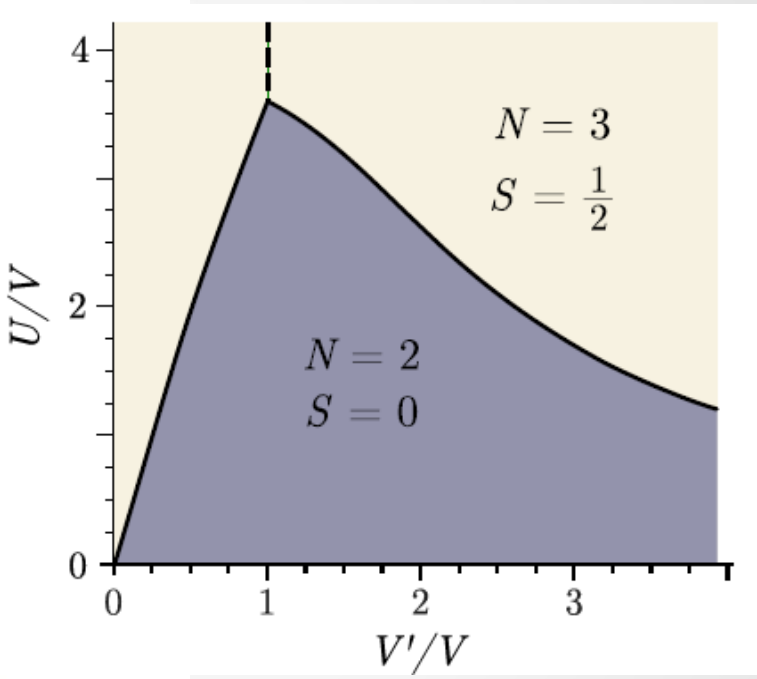
Plan of the talk

- Metastability effects in open correlated systems: going beyond Lindblad approximation
- Emergence of temperature in an emissive correlated system
- Realistic LDA+DMFT calculations of ArCrS2 multiferroic properties

Metastable dynamics: the model





Dynamics: about the Lindblad approximation

Master equation for a Markovian bath

$$\frac{d}{dt}\rho_S(t) = -i[H_S, \rho_S(t)] + \mathcal{L}\rho_S(t)$$

The Lindblad form for the relaxation term:

- is valid in the leading order with coupling constant
- preserves sum rules
- contain creation-annihilation operators of only the site coupled to the bath

We found that Lidblad approximation is valid either for a non-correlated system, or for bath obeying infinite temperature/chemical potential.

Dynamics: beyond the Lindblad approximation

In the generalized theory the relaxation term in the master equation

$$\frac{d}{dt}\rho_S(t) = -i[H_S,\rho_S(t)] + \frac{1}{2}\sum_{\alpha\beta} J_{\alpha\beta}\left(c_\alpha^\dagger \rho_S(t) L_\beta + \right.$$

$$\left. + \bar{L}_\beta \rho_S(t) c_\alpha^\dagger - c_\alpha^\dagger \bar{L}_\beta \rho_S(t) - \rho_S(t) L_\beta c_\alpha^\dagger\right) + \text{h.c.}$$

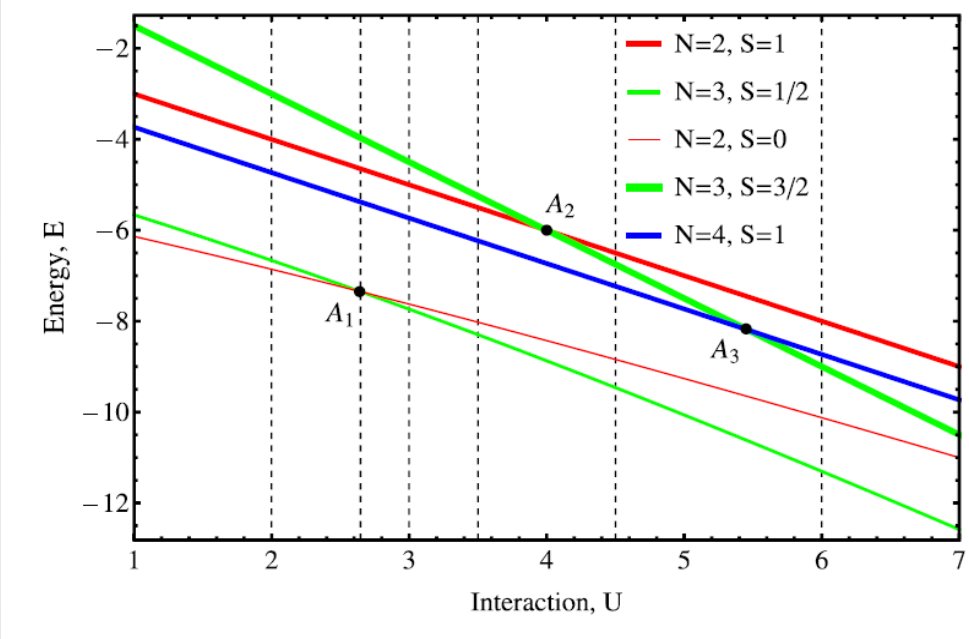
has a many-body nature (sum is over many-body states, *f* is Fermi function)

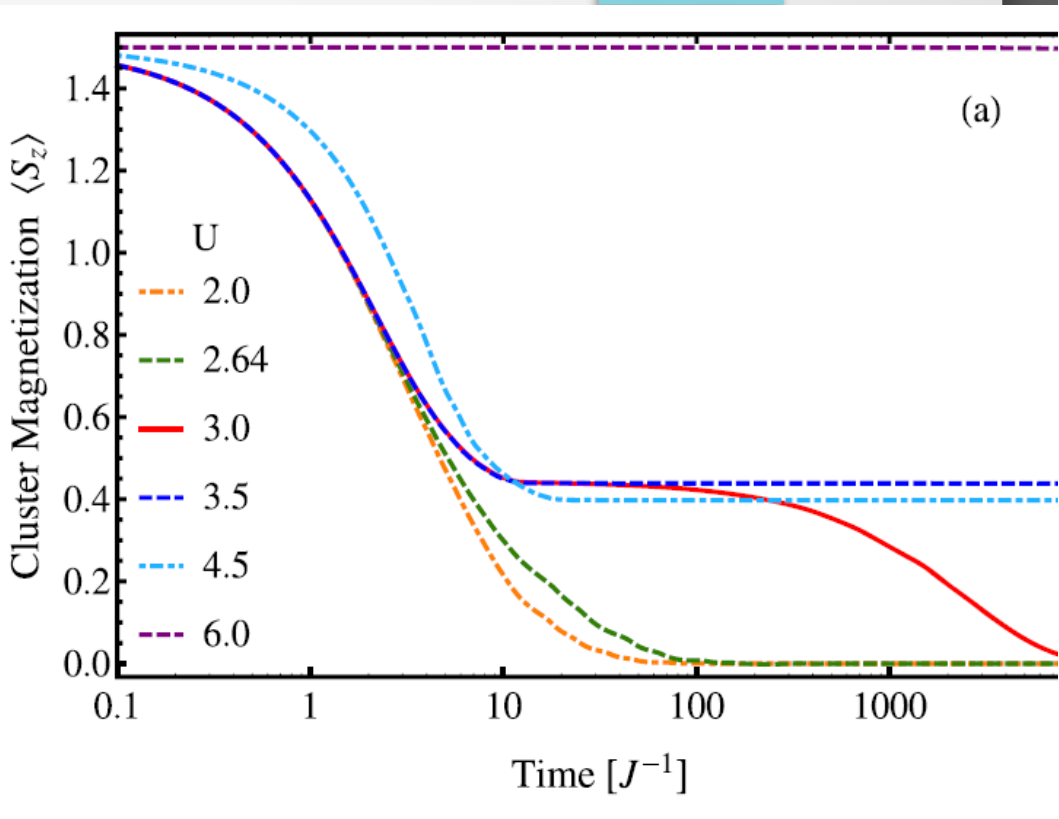
$$L_\alpha = \sum_{mn} f(\varepsilon_{nm}) \langle m|c_\alpha|n\rangle |m\rangle\langle n|$$

$$\bar{L}_\alpha = \sum_{mn} (1 - f(\varepsilon_{nm})) \langle m|c_\alpha|n\rangle |m\rangle\langle n|$$

The formalizm passes into Lindblad assuming *f*=const.

Metastable dynamics: the results



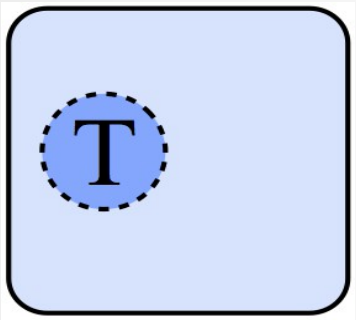


Metastability occurs due to selecton rules for the transitions between many-body states

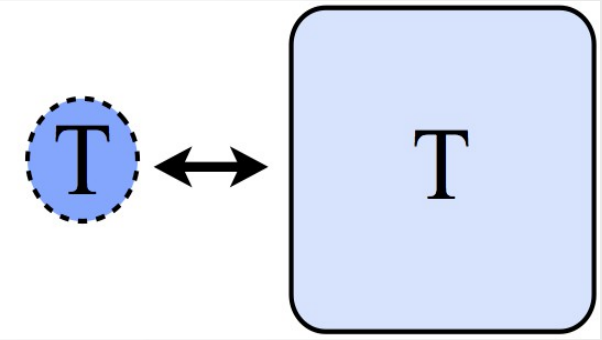
Calculus for realistic systems (say, surface cathalisys) will require huge matrices

The concept of temperature

Isolated systems



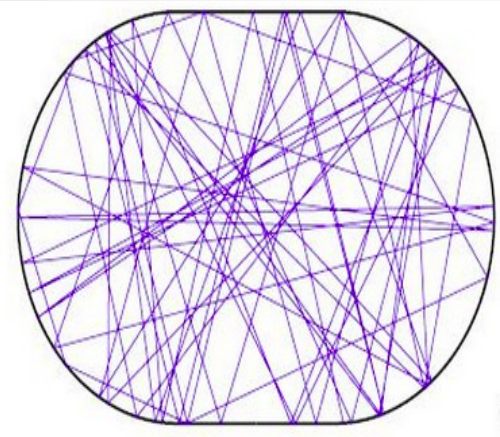
Open systems



Macroscopic limit

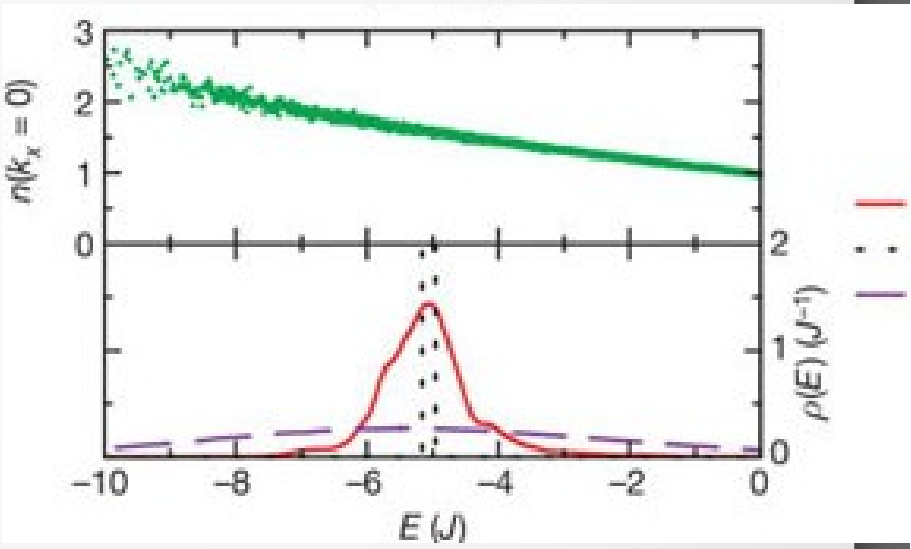
Thermalization of isolated systems

Classical systems:
ergodic hypotesis



All phase space available
(trajectory is a microcanonical ensemble)

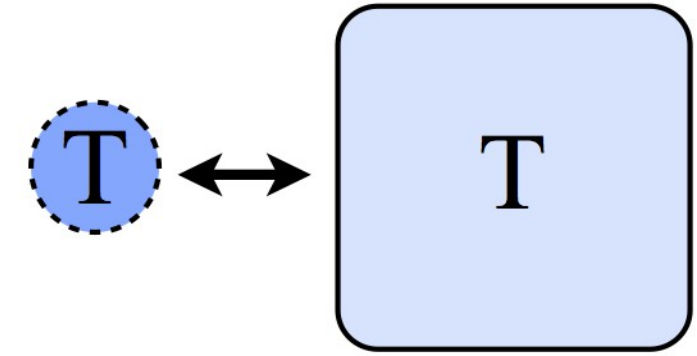
Quantum systems:
Eigenstate thermalization hypotesys



Observables almost do not vary between eigenstates close in energy
(eigenstate is a microcanonical ensemble)

M. Rigol et al. Nature 452, 854 (2008)

Thermalization of open systems

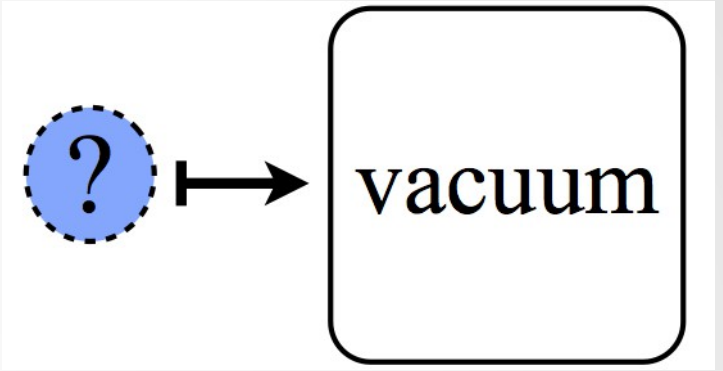


Detailed balance principle

$$r_{m\to n}e^{-E_m/T} = r_{n\to m}e^{-E_n/T}$$

Thermalization without detailed balance

Macroscopic limit: averages

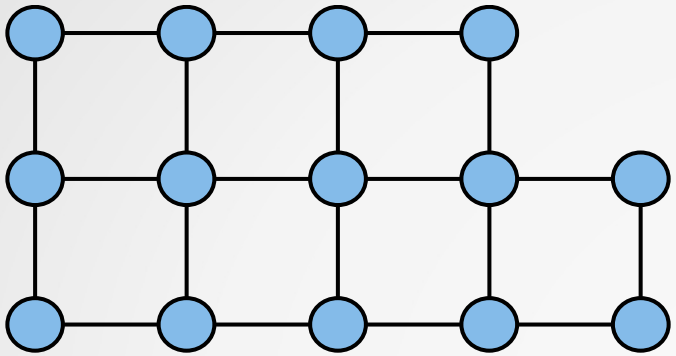


emissive system

Finite-size: Boltzmann (???) distribution of populations

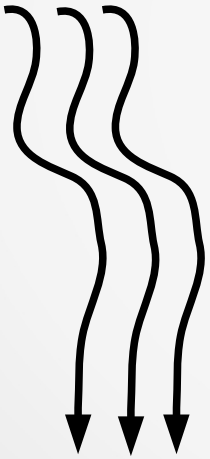
Hard-core bosons on a lattice with emission

System



$$H_S = -\sum_{\langle ij \rangle} h_{ij}(b_i^\dagger b_j + b_j^\dagger b_i)$$

Constraint: each site is occupied no more then once



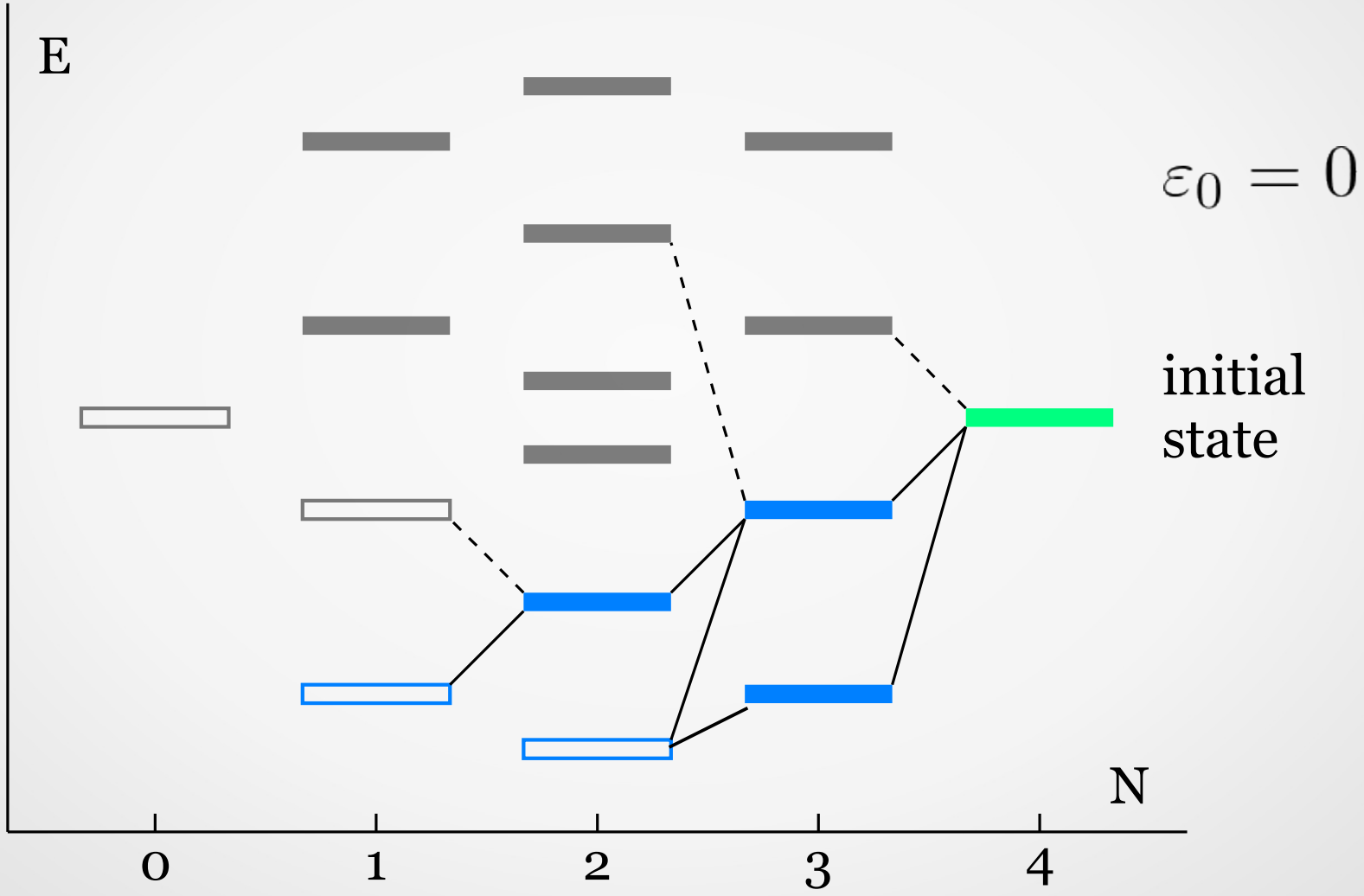
Coupling

$$H_R = \sum_k \varepsilon_k a_k^\dagger a_k$$

Reservoir

$$H_I = \alpha \sum_{ki} (a_k^\dagger b_i + b_i^\dagger a_k)\theta(\varepsilon_k - \varepsilon_0)$$

The evolution of a system at 4-site lattice



Coarse-grained master equation

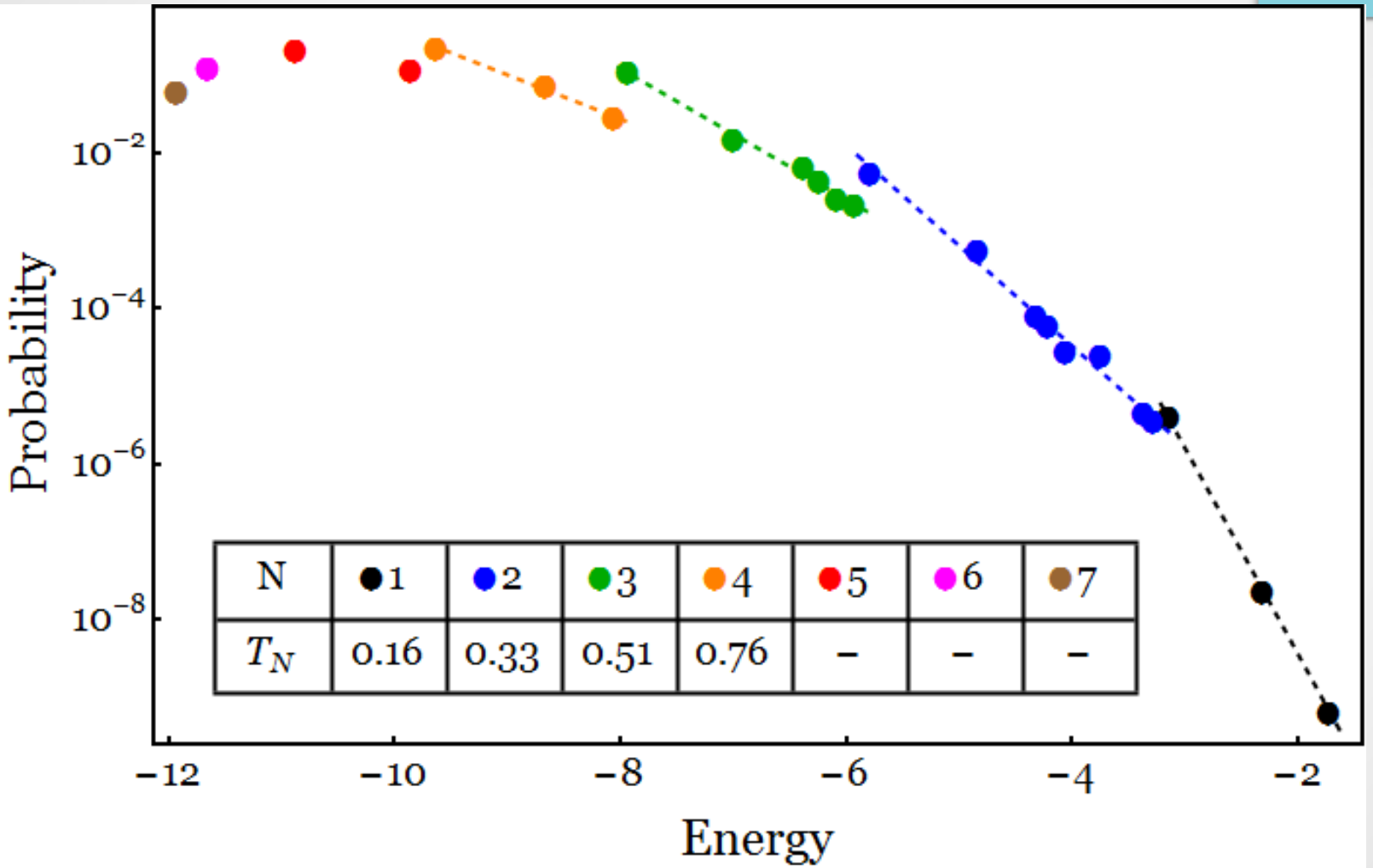
$$\frac{d}{dt}P_n^N = \sum_m R_{nm}^{N+1} P_m^{N+1} - \sum_m R_{mn}^N P_n^N$$

$$\langle N, n|\rho_S|N, n\rangle \equiv P_n^N$$

Fermi's golden rule

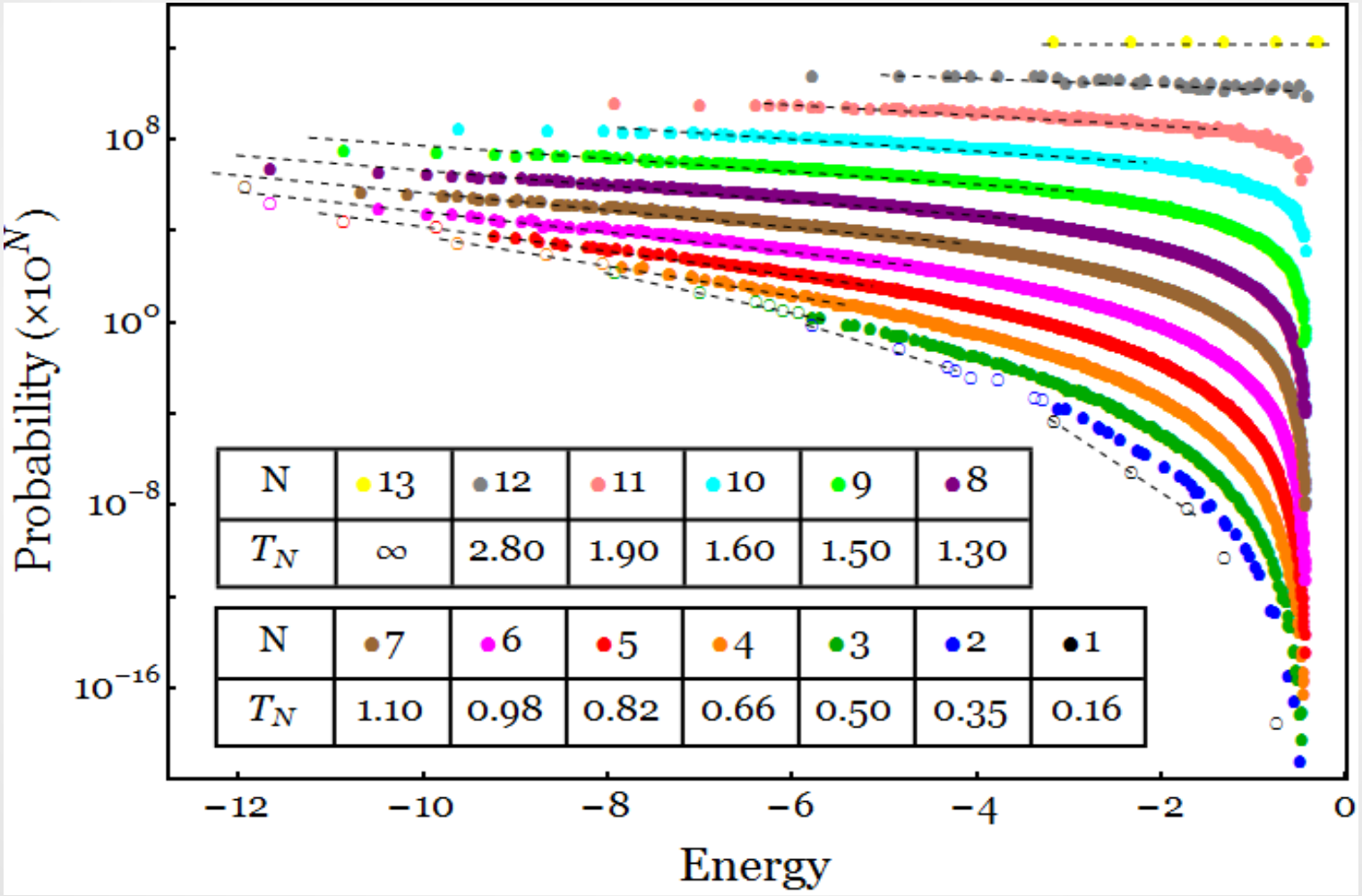
$$R_{mn}^N = 2\pi\Omega_0\alpha^2 \sum_{i=1}^{L} |\langle N-1, m|\hat{b}_i|N, n\rangle|^2$$

Distribution over stable states

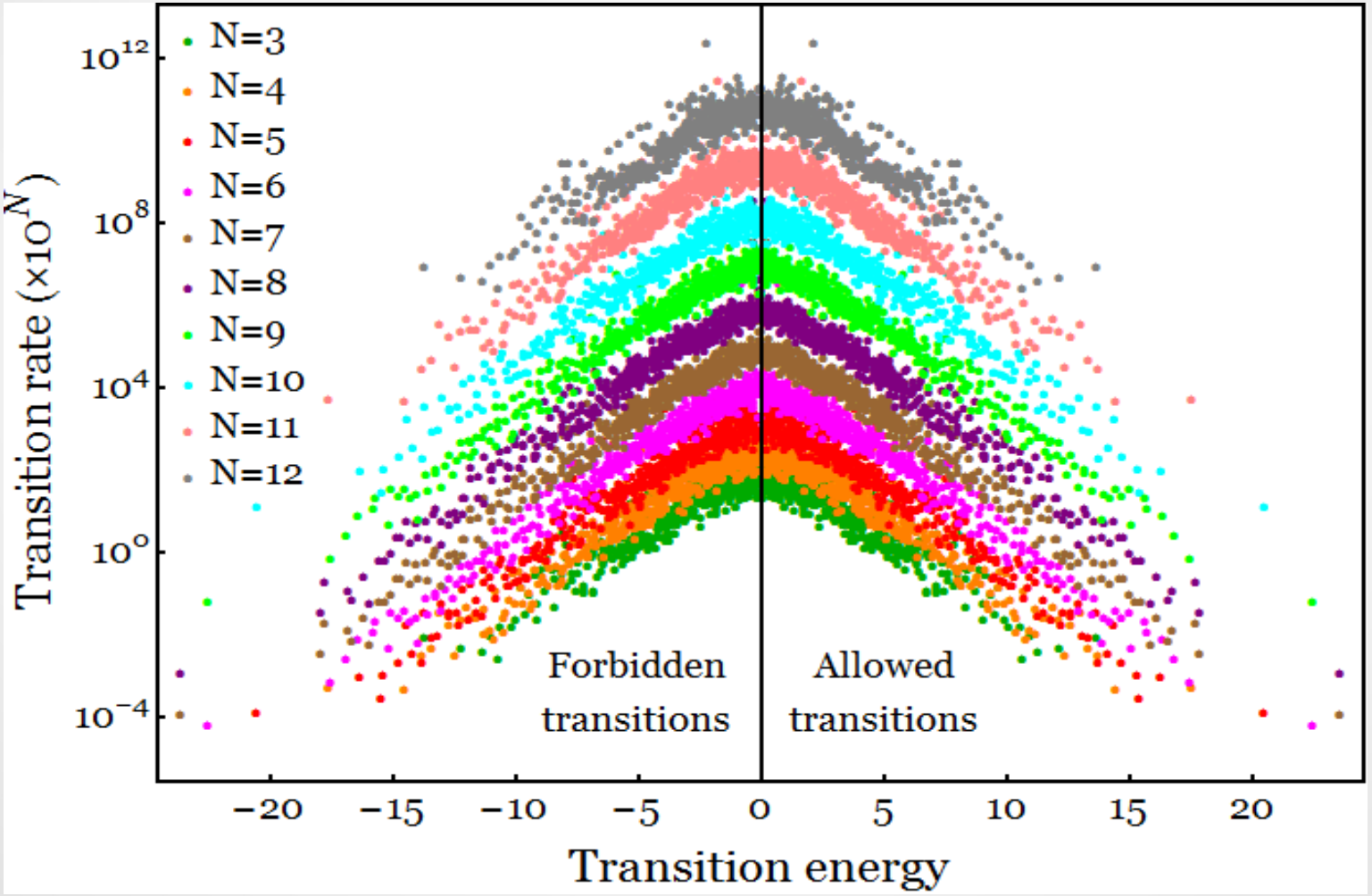


Bolzmann statistics in the sectors with same particle numbers

Distribution over intermediate states

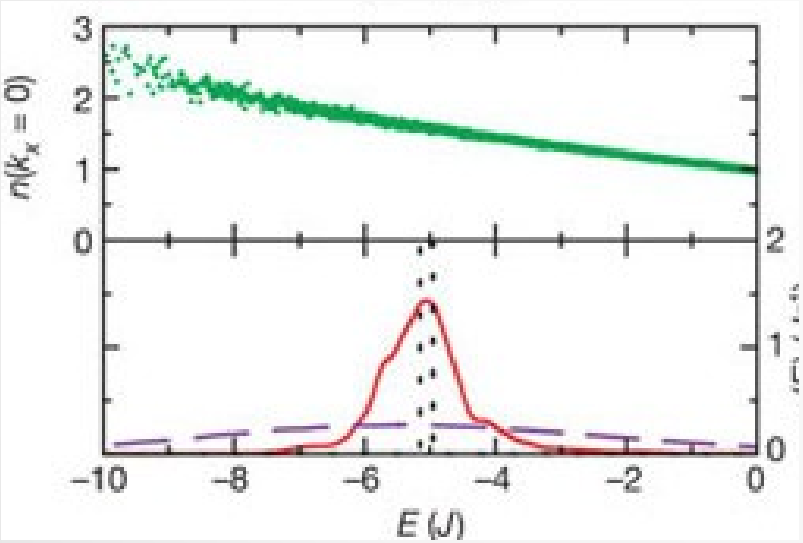


Transition rate vs. transition energy

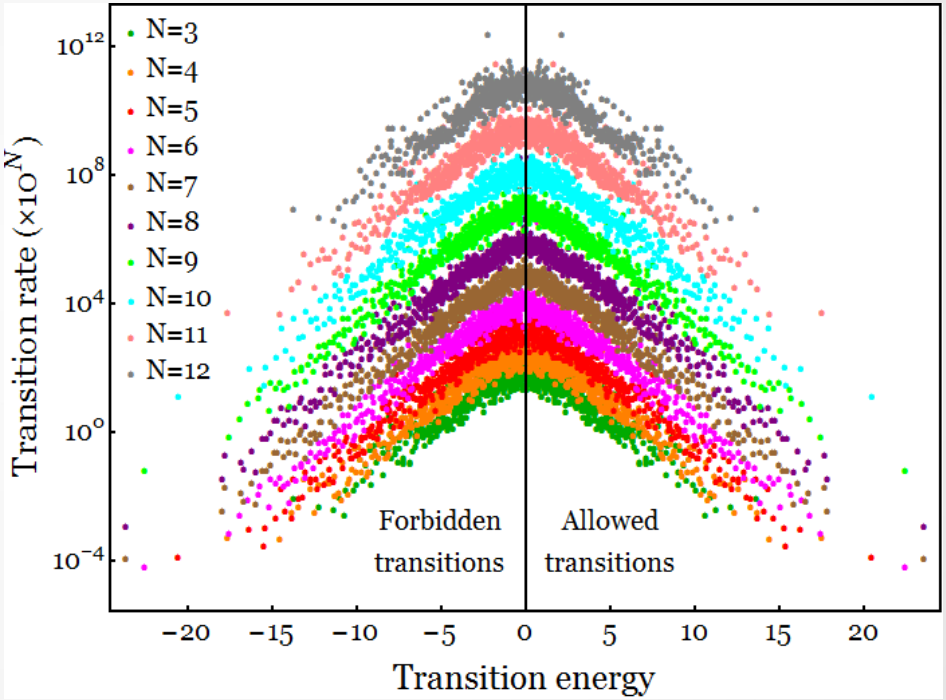


Roots of the "temperature"

Isolated quantum systems:
Eigenstate thermalization hypotesys



Emissive quantum systems:
This work



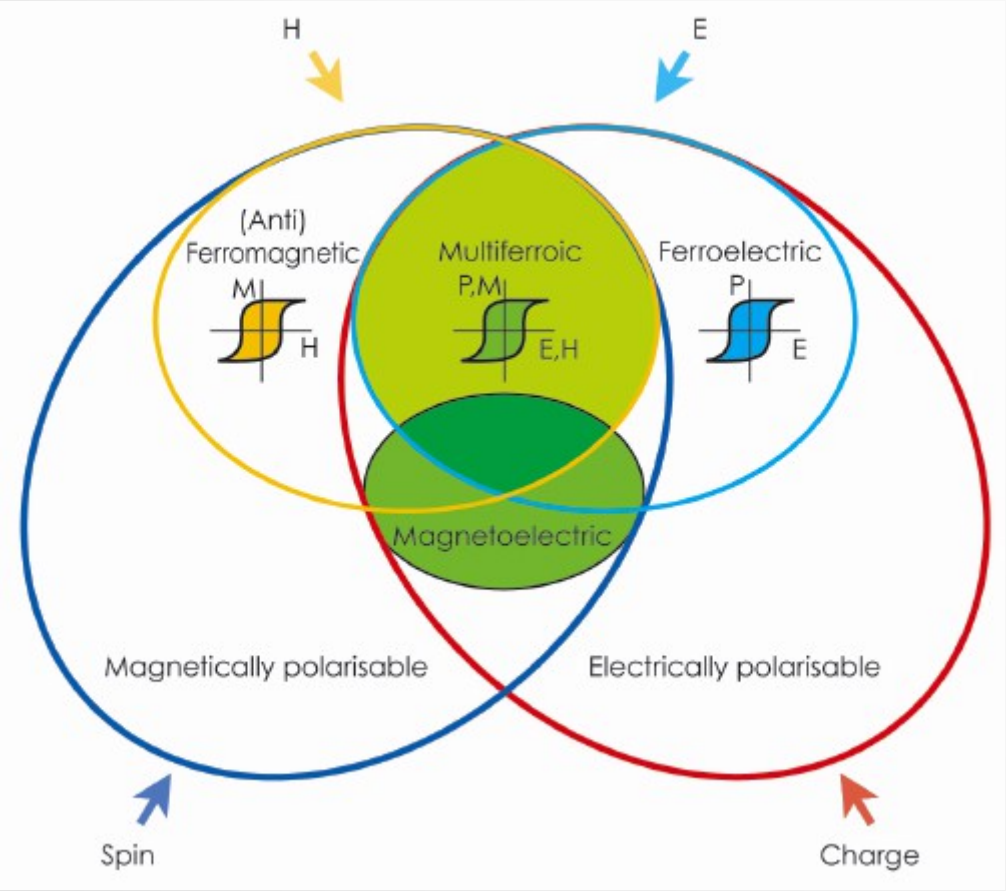
Each eigenstate itself forms
a microcanonical ensemble

Emissing a particle from an eigenstate
forms a canonical ensemble

Working with soft-core bosons will require huge matrices

Multiferroics

Multiferroics exhibit more than one primary ferroic order parameter simultaneously

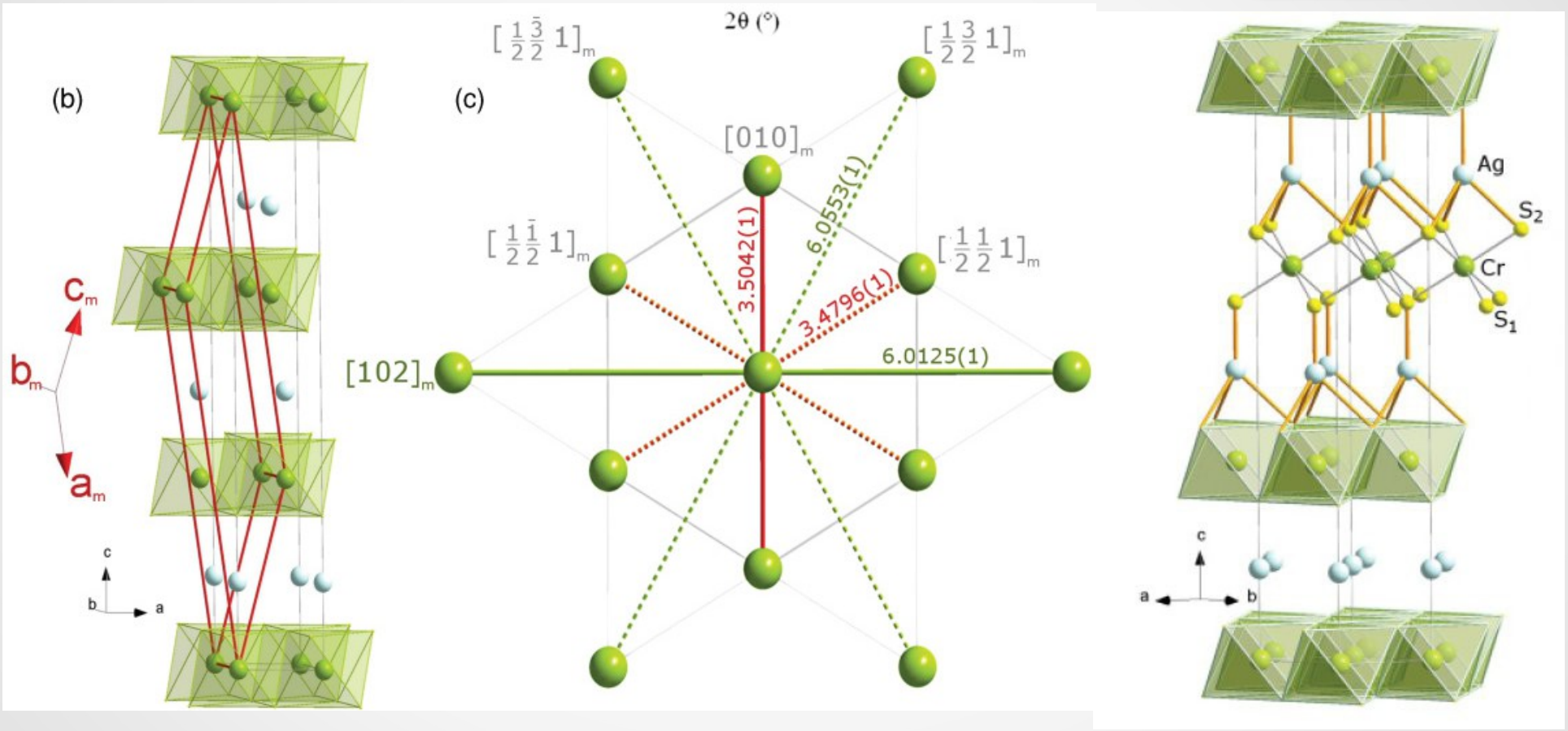


- ✓ Ferromagnetism
- ✓ Ferroelectricity
- ✓ Ferroelasticity
- ✓ Ferrotoroidicity

Crystal structure of $AgCrS_2$

Low-T (Cm)

High-T (R3m)

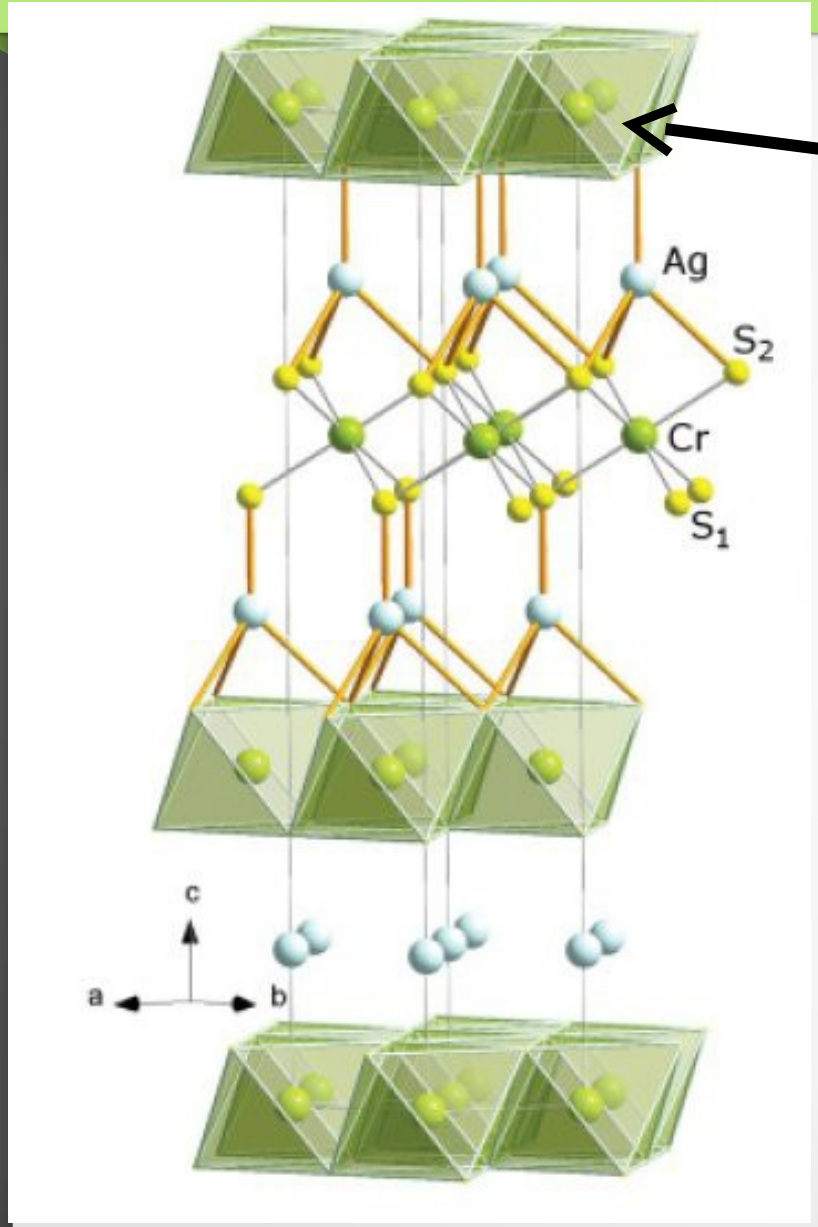


Layered structure with triangular topology and two phase transitions at 670 K and 41 K

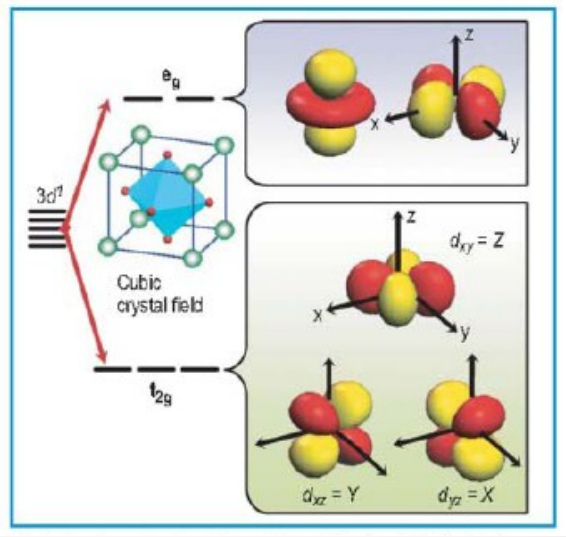
from R-3m via R3m to Cm

Damay et al., Phys. Rev. B **83**, 184413 (2011

Local environment of Cr ions



Cr ions are surrounded by octahedrons of sulfur

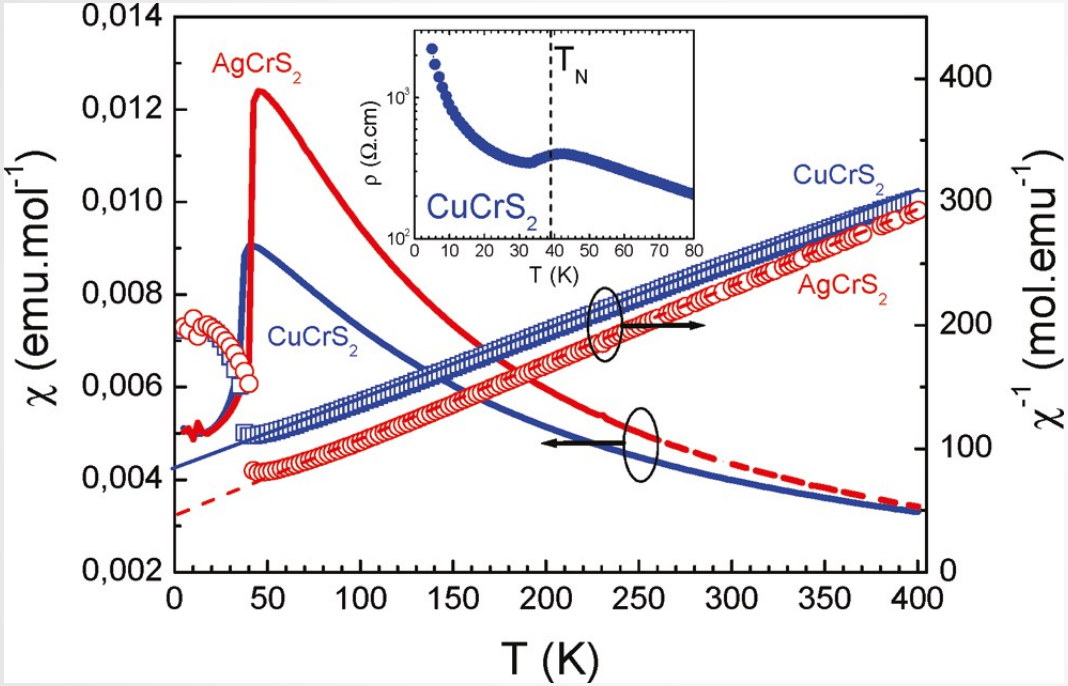


$Ag^{1+}Cr^{3+}S^{2-}_{2}$

3 electrons in degenerate t_{2g} subshell

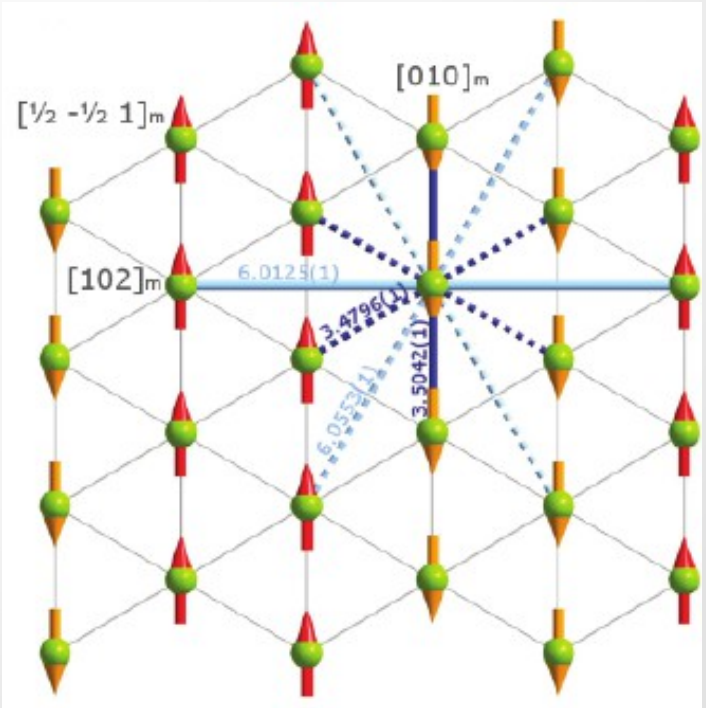
Magnetic properties

Effective moment value confirms 3 electrons in t_{2g} subshell !!!



T_N = 42 K

μ = 2.7 μ_B

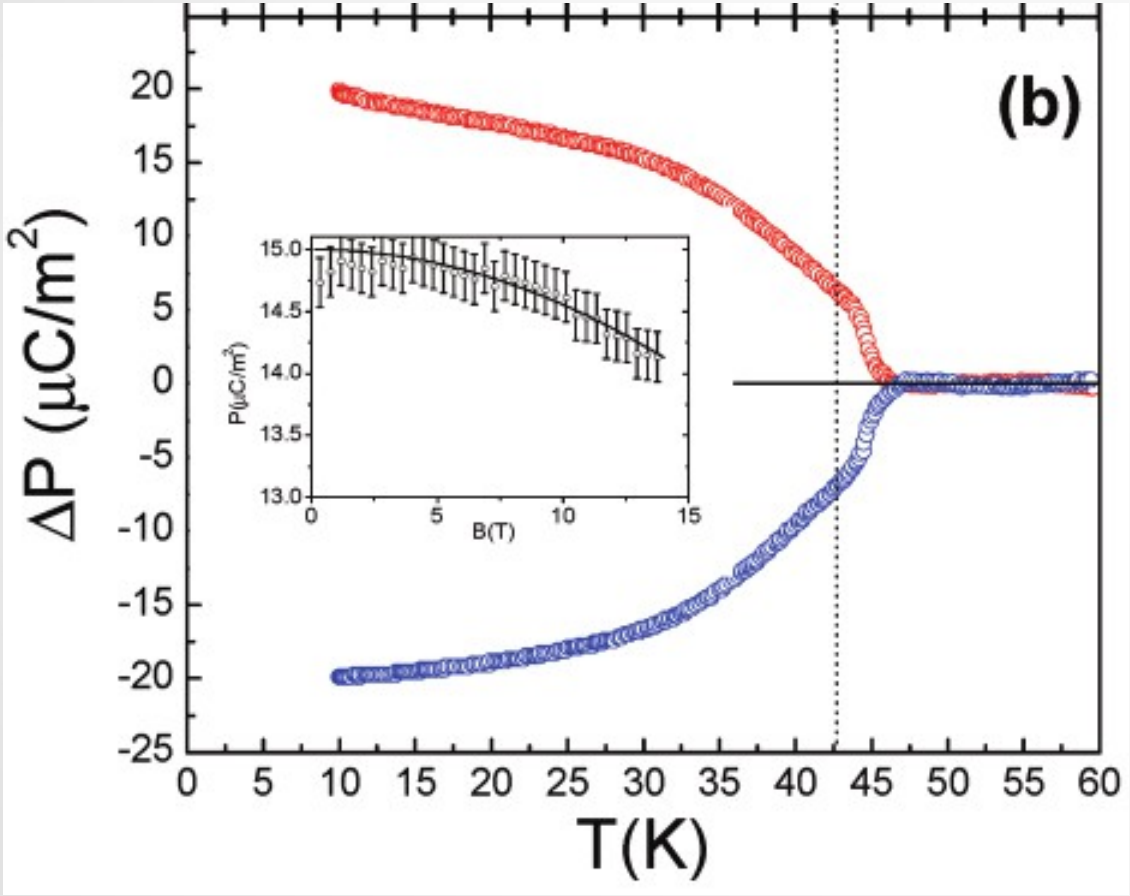


Unusual magnetic structure with double stripes!!!

Damay et al., Phys. Rev. B **83**, 184413 (2011)

Low temperature electrical properties of $AgCrS_2$

Electric polarization



Multiferroic below 42 K
with $\Delta P = 20\ \mu C/m^2$

Singh et al., Chem. Mater. **21**, 5007 (2009)

First principles calculations

Berry phase formalism to calculate polarization

GGA

	FM→4S
$\delta\vec{P}$, eÅ	[0.239; 0.059; -0.127]
$\lvert\delta P\rvert$, eÅ	0.277
$\lvert\delta P\rvert$, $\mu C/m^2$	7756

Experimental value
$|\delta P| = 20\ \mu C/m^2$

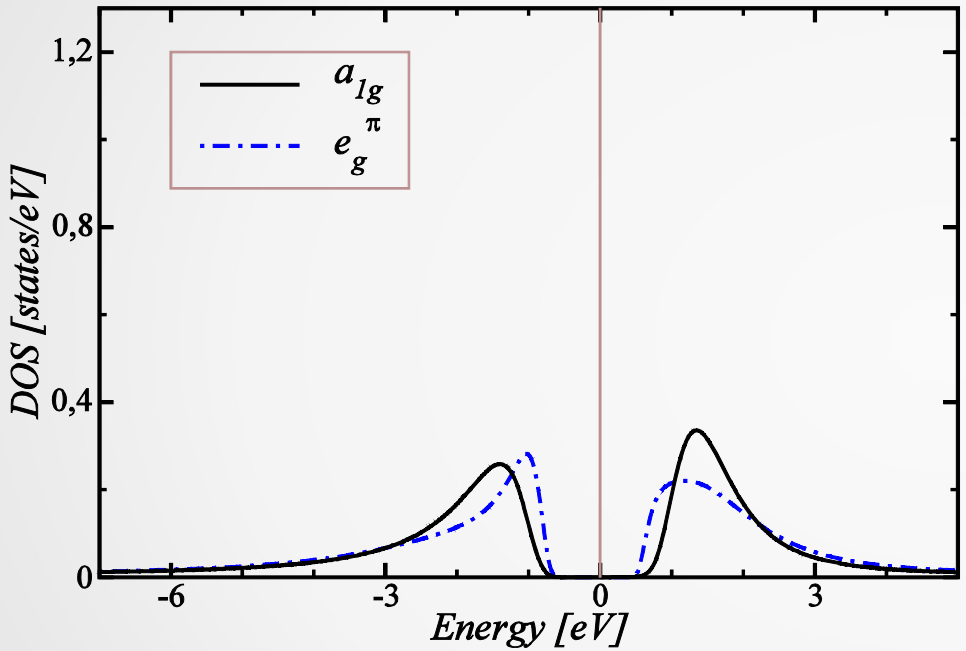
GGA+U

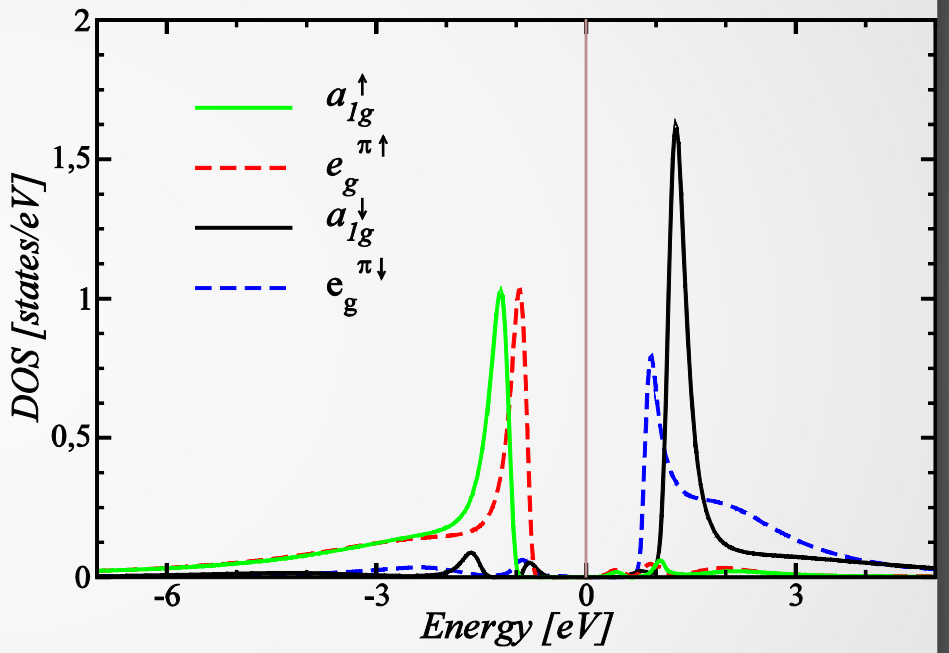
	FM→4S	AFM→4S
$\delta\vec{P}$, eÅ	(-0.070; -0.018; 0.037)	(-0.077; -0.019; 0.054)
$\lvert\delta P\rvert$, eÅ	0.08	0.10
$\lvert\delta P\rvert$, $\mu C/m^2$	2240	2660

Huge overestimation of polarization (2 orders of magnitude)!!!
Clear importance of correlations
Starting point insensitive (both wrong)

Simple model

R3m structure t_{2g} electrons





PM insulator

Magnetic insulator

$\mu = 2.4\ \mu_B$

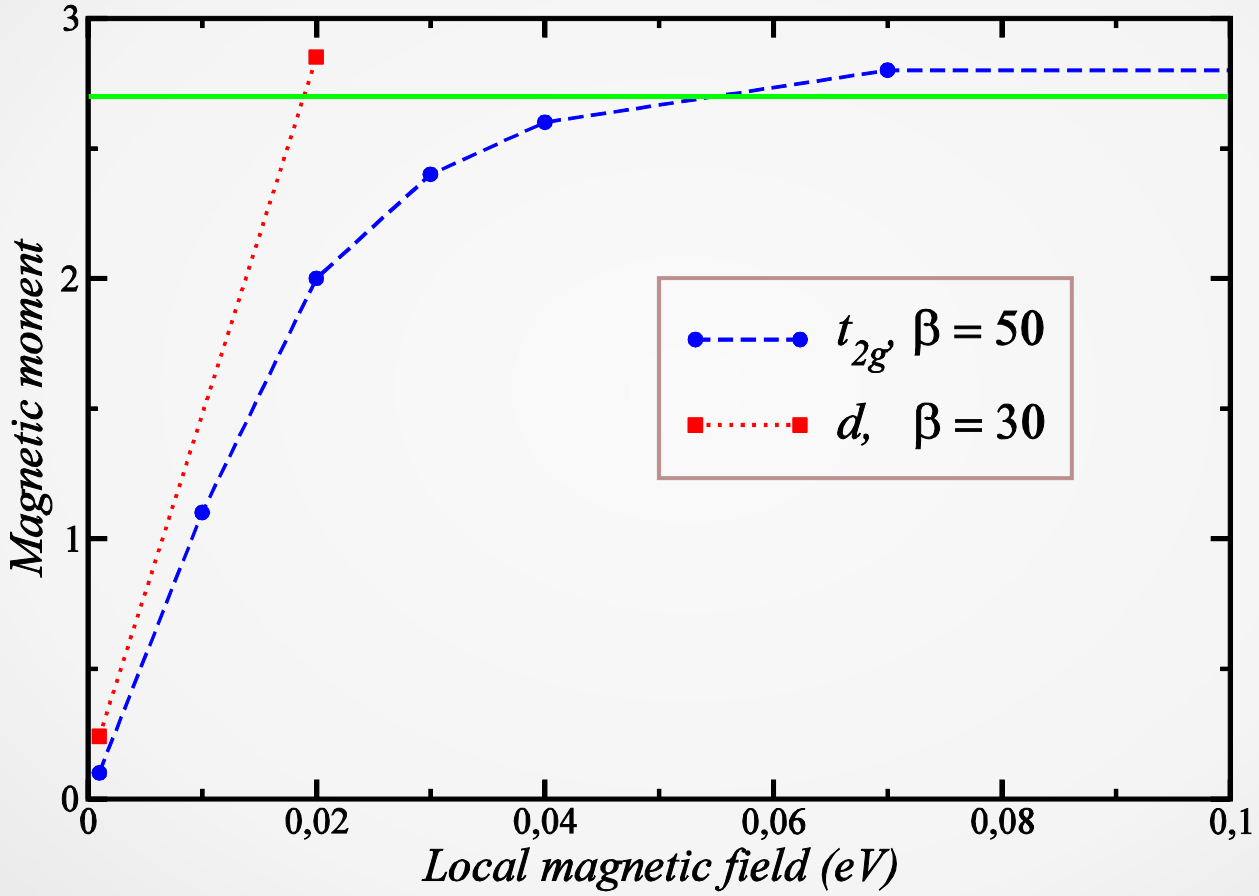
Calculated value for electric polarization $|\delta P| = 103\ \mu C/m^2$

Polarization for strongly correlated systems

- Nourafkan & Kotliar formalism

$$\Delta \mathbf{P} = -\int_0^1 d\xi \left(\frac{ie}{2N\beta}\right) \sum_{\mathbf{k}\sigma} \sum_m Tr \left(\frac{\partial \hat{G}^{-1}}{\partial \mathbf{k}} \hat{G} \frac{\partial \hat{G}^{-1}}{\partial i\omega_m} \hat{G} \frac{\partial \hat{G}^{-1}}{\partial \xi} \hat{G} - \frac{\partial \hat{G}^{-1}}{\partial \mathbf{k}} \hat{G} \frac{\partial \hat{G}^{-1}}{\partial \xi} \hat{G} \frac{\partial \hat{G}^{-1}}{\partial i\omega_m} \hat{G} \right)$$

Local magnetic moment of $AgCrS_2$ in LDA+DMFT



Strong dependence on the orbital degeneracy!!!

Realistic material calculations

or how it should be done for $AgCrS_2$

- 8 f.u. in correct 4SL magnetic structure
- Accurate band structure calculations along R3m-Cm path (about 5 points)
- LDA+DMFT calculation at each structural point
- Nourafkan & Kotliar formalism to calculate electric polarization

All steps are time consuming!!!

DMFT example with the same “technical” parameters and 128 procs

- 3-bands, β=30 - 12 mins (time per iteration)
- 3-bands, β=50 - 20 mins
- 5-bands, β=30 - 70 mins