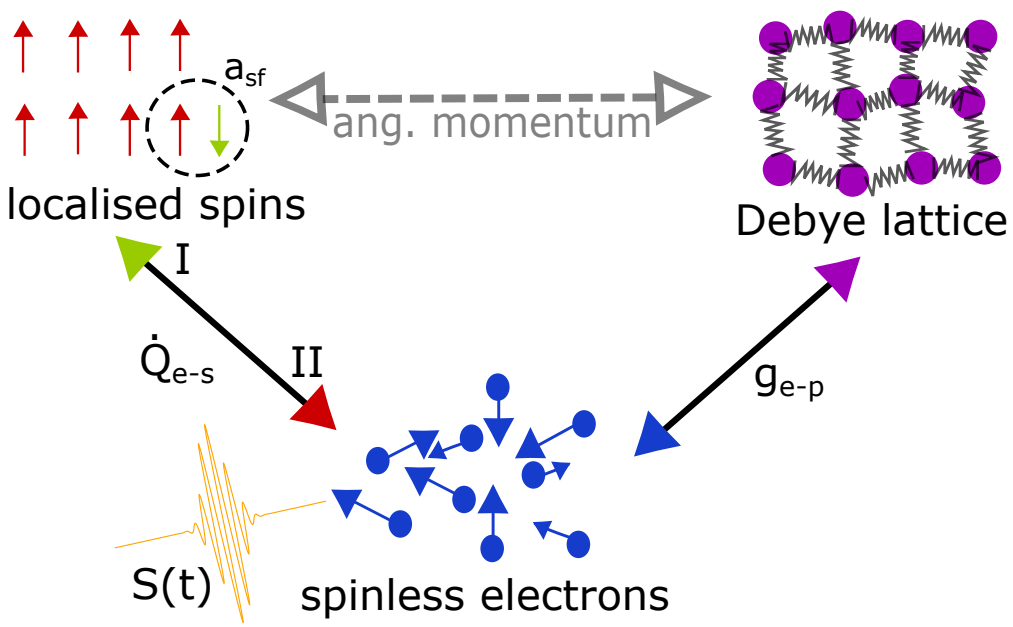
M3TM model concept



- decoupled systems of spinless electrons, phonons and localized spins in Mean field approximation (MFA)
- laser pulse heats electron system
- energy distribution to lattice and spins
  (for FGT: no remagnetization so energy flow $\dot{Q}_{es}$ (see next slide) unidirectional e $\rightarrow$ s)
- spin flips (/magnon excitations) upon e-p scattering event from Elliott-Yafet spin mixing
- implicit angular momentum exchange between spins and lattice

M3TM equations

Dynamics of electronic, lattice and spin subsystems[6]

$$C_{\mathrm{e}} \frac{dT_{\mathrm{e}}}{dt} = g_{\mathrm{e-p}}(T_{\mathrm{p}} - T_{\mathrm{e}}) + S_0\, G(t,z)) + \dot{Q}_{\mathrm{es}}$$

$$C_{\mathrm{p}} \frac{dT_{\mathrm{p}}}{dt} = -g_{\mathrm{e-p}}(T_{\mathrm{p}} - T_{\mathrm{e}})$$

$$\frac{dm}{dt} = Rm\frac{T_{\mathrm{p}}}{T_C}\left(1 - \frac{m}{B_S\left(\frac{Jm}{k_B T_e}\right)}\right)$$

$C_{e(p)}$ electron (phonon) heat capacity

g_{ep} electron phonon coupling

$\dot{Q}_{es} = Jm\dot{m}/V_{at}$ energy exchange of spin-and electron system

$\mathrm{R} = 8\frac{a_{sf}\, g_{ep}\, T_C\, V_{at}}{\mu_{at}\, k_B\, T_{Deb}^2}$ magnetization rate parameter

a_{sf} spin flip probability upon e-p-scattering event

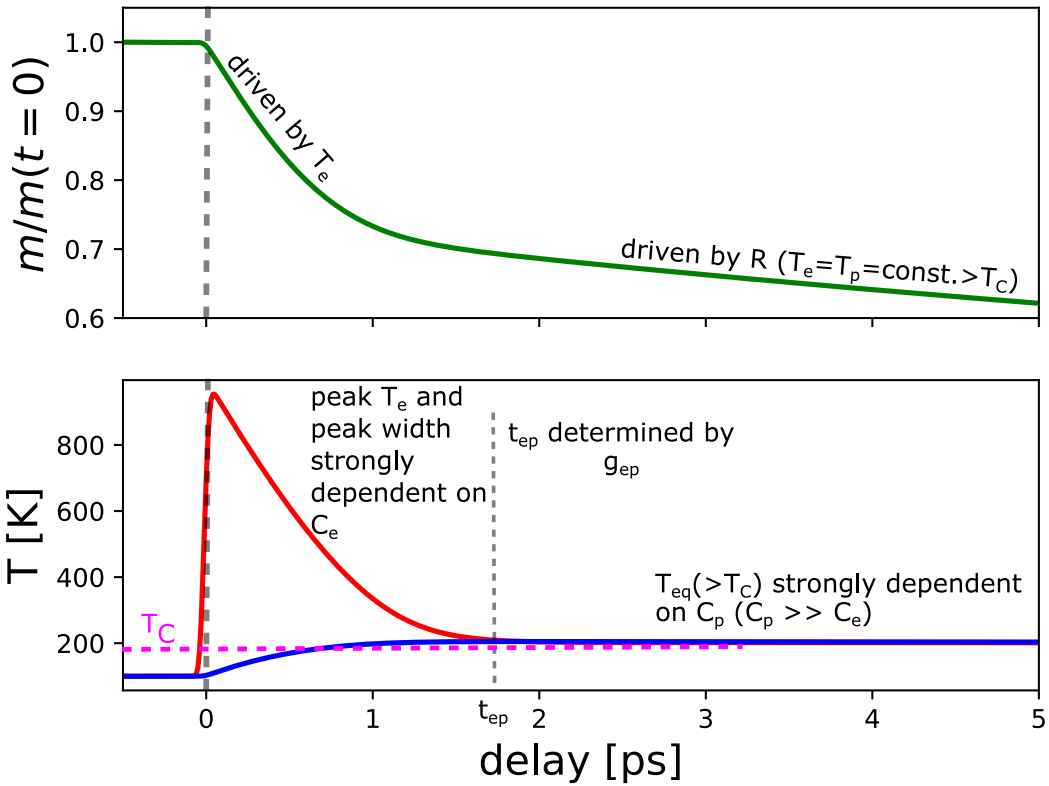
B_S Brillouin function for effective spin S

parameter impact

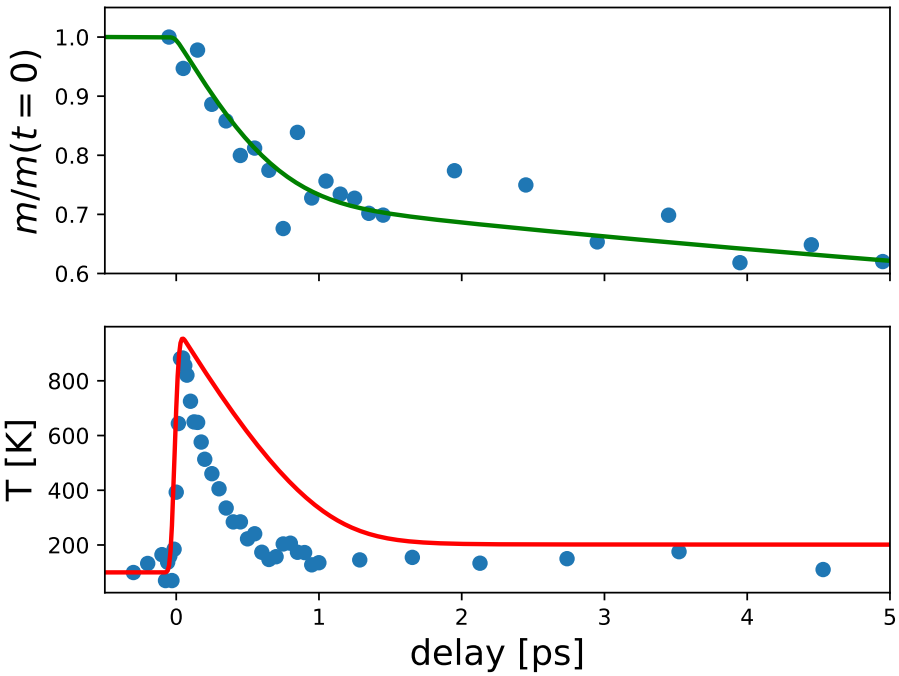
easy first attempt: take parameters used before for a very similar model

Model	symbol	meaning	estimate	unit
M3TM	$\mu_a t$	Atomic magnetic moment [a]	1.47×10^{-23}	Am^2
	γ	Electron heat capacity constant [a]	1561	$Jm^{-3}K^{-2}$
	V_{at}	Atom spin density volume [a]	1.68×10^{-29}	m^3
	T_C	Curie temperature [b]	191	K
	σ	pulse width [b]	85	fs
	g_{ep}	Electron-phonon relaxation rate [c]	1.33×10^{6}	$Jm^{-3}K^{-1}ps^{-1}$
	C_p	phonon heat capacity [c]	6.28×10^{6}	$Jm^{-3}K^{-1}$
	E_D	Debeye energy [c]	6.49×10^{-21}	J
	a_{sf}	Spin-flip probability	0.14	unity

Lichtenberg et al., arXiv:2206.01452v1

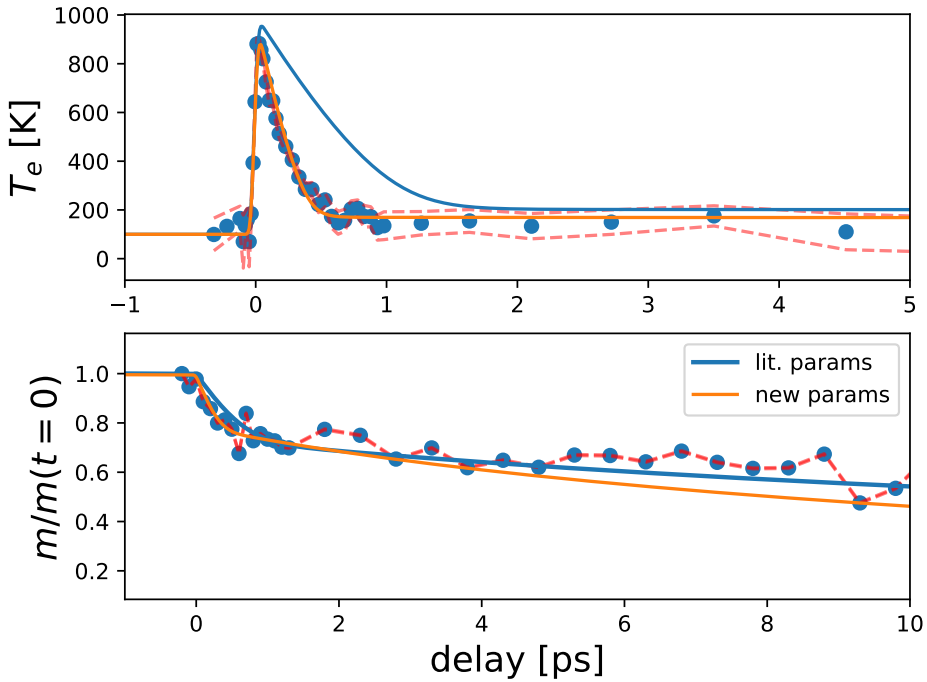


Fitting with literature parameters



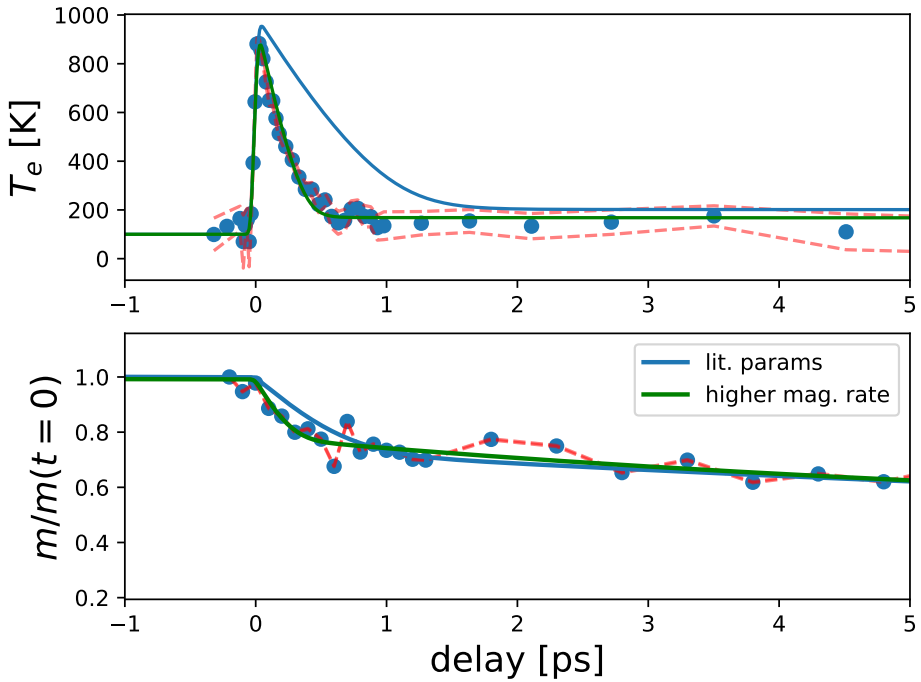
- parameters used to fit magnetization dynamics in paper cited above
- good fit of mag dynamics
- electron dynamics $\approx$ 3 times slower than in experiment
- equilibrium temperature too high in simulations

first try of fit adjustment



- increase c_p by a factor of 1.5 to adjust equilibrium magnetization
- increase $g_e p$ by a factor of 3 to speed up electron dynamics
- decrease spin flip probability by a factor of 3 to stabilize magnetization dynamcis
- $\rightarrow$ **electron dynamics well reproduced**
- **magnetization rate after e-p-equilibration seems to fast**
- **next try: increase a_{sf} at the cost of pump power to match the magnetization behaviour for $t > \tau_{ep}$**

Fitting irrespective of absorbed energies



- both T_e and m are nicely reproduced
- lattice dynamics data important to validate use of g_{ep}, c_p
- instead of rate parameter, also c_e can be changed to adapt peak of T_e and magnetization rate
- investigation for multiple fluences important (I'd like to talk again about the initial consitions and repetition rate/effective starting temperature)

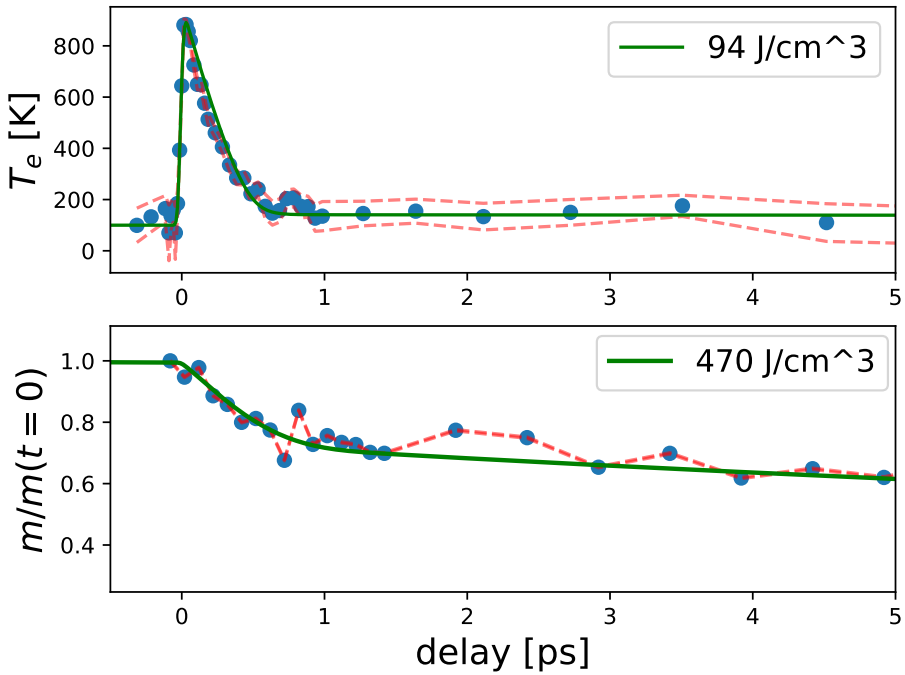
New fits respecting absorbed energies

- treating experimental absorbed energies:
  - XMCD/UED signals recorded in transmission, so average absorbed energy density can be directly used for monolayer simulations
  - ARPES signal detected from hot surface of the sample, so that simulated absorbed energy adjusted by
  - $E_{sim} = E_{exp}\ N/(\sum_{i=0}^{N} \exp\left(-\frac{idz}{\lambda}\right))$
  - dz is unit cell thickness, λ is penetration depth of laser pulse, for $N = 1:\ E_{sim} = E_{exp}$

Absorbed energy density $[\frac{J}{cm^3}]$	measurement
98	ARPES
250-380	UED
490	XMCD

New fits respecting absorbed energies

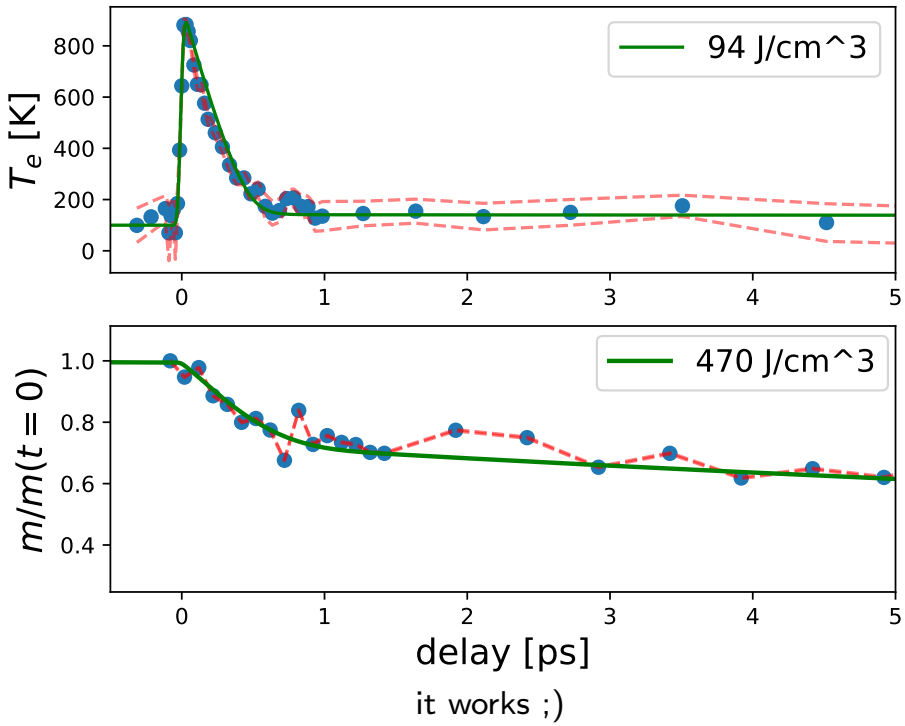
- fix absorbed energy for ARPES signal, then adjust C_e, g_{ep} and C_p to fit data
- increase pump power by factor of 5 and hope that adjusting spin-flip probability a_{sf} (mag. rate parameter R) fits XMCD signal



it works ;)

New fits respecting absorbed energies

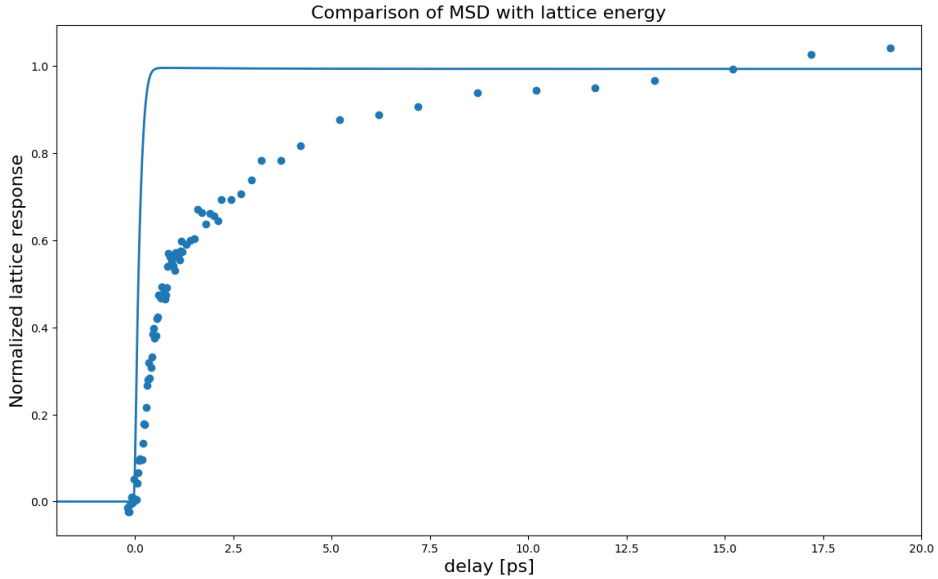
parameter [units]	used	Fe for ref
γ $[\frac{kJ}{m^3K^2}]$	0.2	$\approx$ 0.8
$C_{p,max}$ $[\frac{MJ}{m^3K}]$	4	3.4
g_{ep} $[\frac{EW}{m^3K}]$	0.44	$\approx$ 1.5



it works ;)

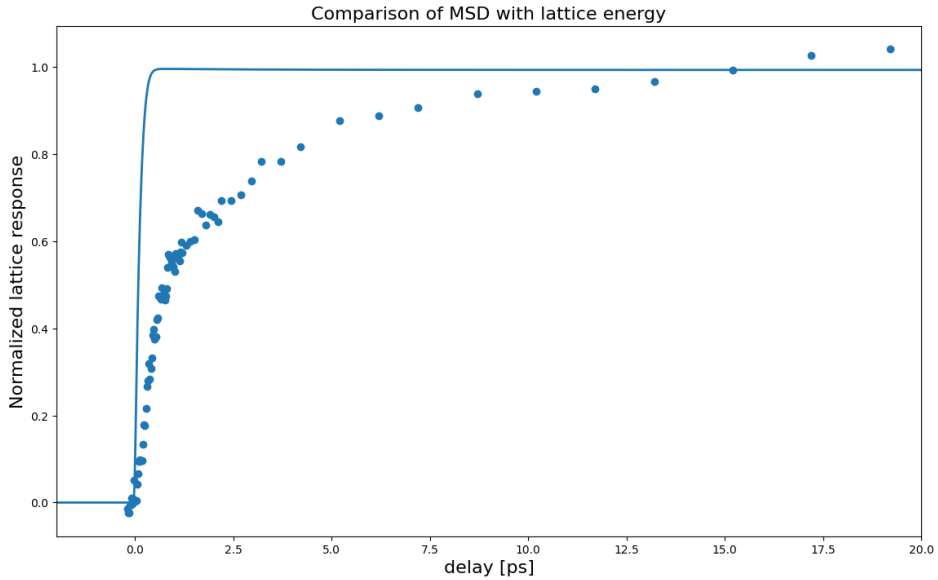
Lattice response

- fitted is norm. $\Delta(C_p T_p)$ to norm. MSD
- above simulation parameters do not fit lattice response



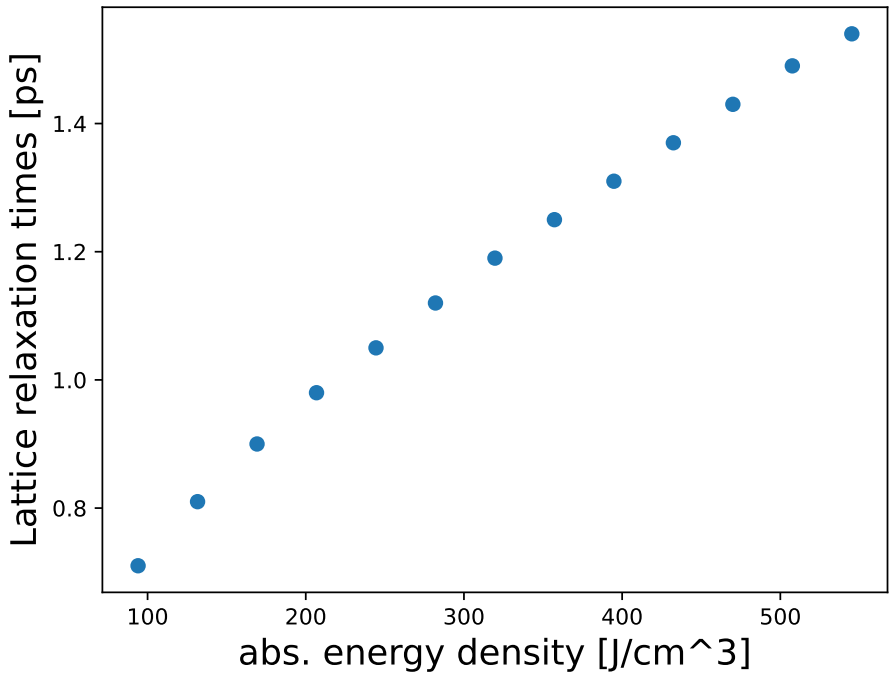
Lattice response

- lattice equilibrates on similar timescale **as electron temperature** ≤ 1 ps
- lattice temperature **descreases** slighty over time due to $\dot{Q}_{\mathrm{es}} < 0$



Lattice relaxation times

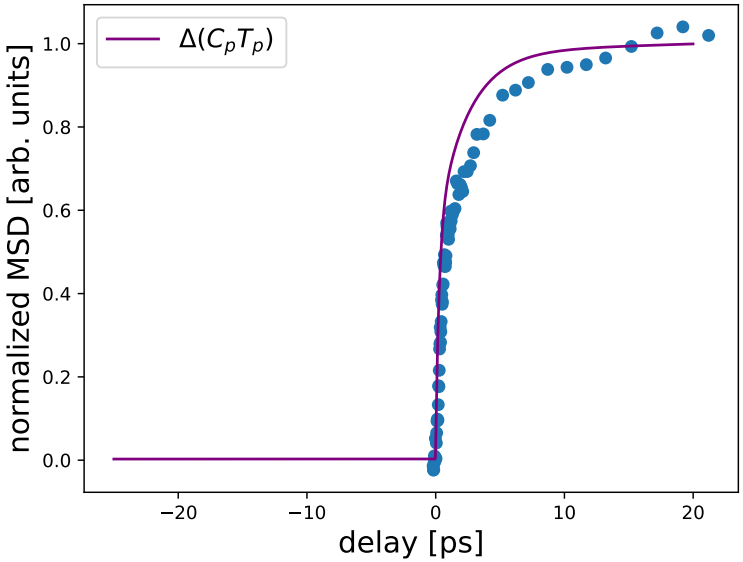
- lattice equilibrates way slower than electrons



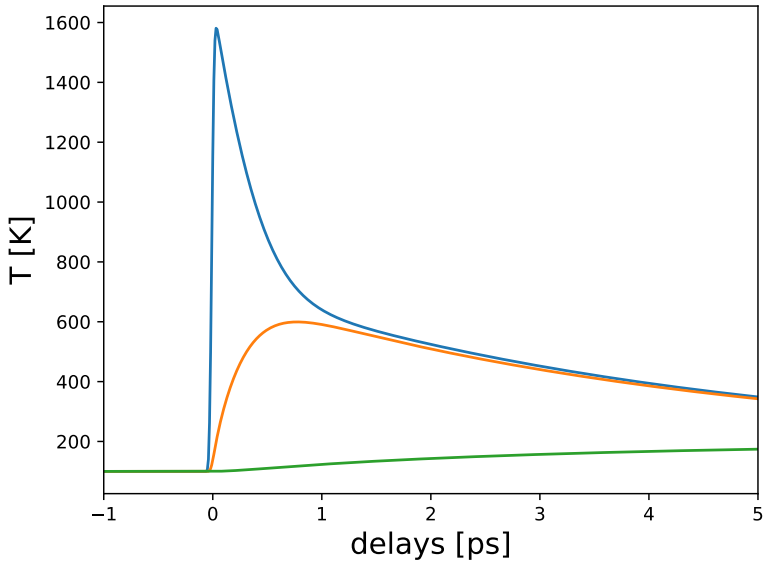
Multi-lattice modeling

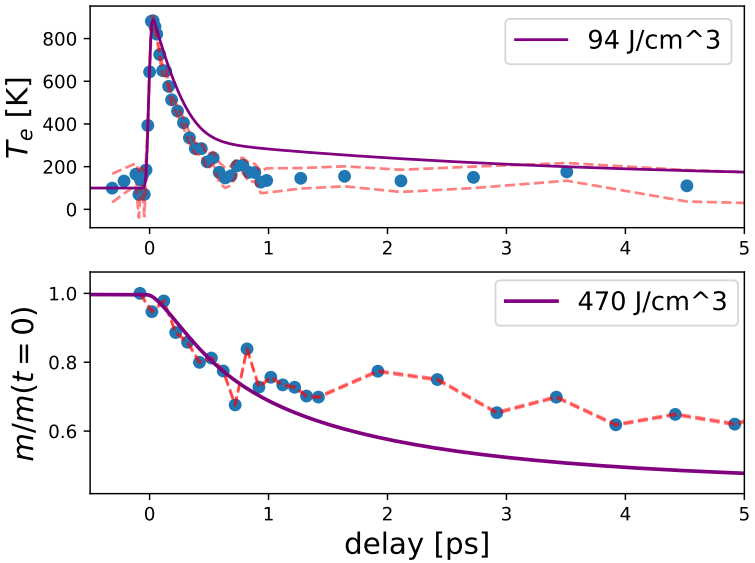
- assume not the full lattice is coupled to electrons
- two subsystems of phonons coupled through phonon-phonon coupling

$$
\begin{aligned}
C_{p,e}\,T_{p,e} &= kC_p(T_{p,e})T_{p,e} \\
C_{p,p}\,T_{p,p} &= (1-k)C_p(T_{p,p})T_{p,p} \\
C_{p,e}\frac{dT_{p,e}}{dt} &= g_{ep}(T_e - T_{p,e}) + g_{pp}(T_{p,p} - T_{p,e}) \\
C_{p,p}\frac{dT_{p,p}}{dt} &= g_{pp}(T_{p,e} - T_{p,p})
\end{aligned}
$$



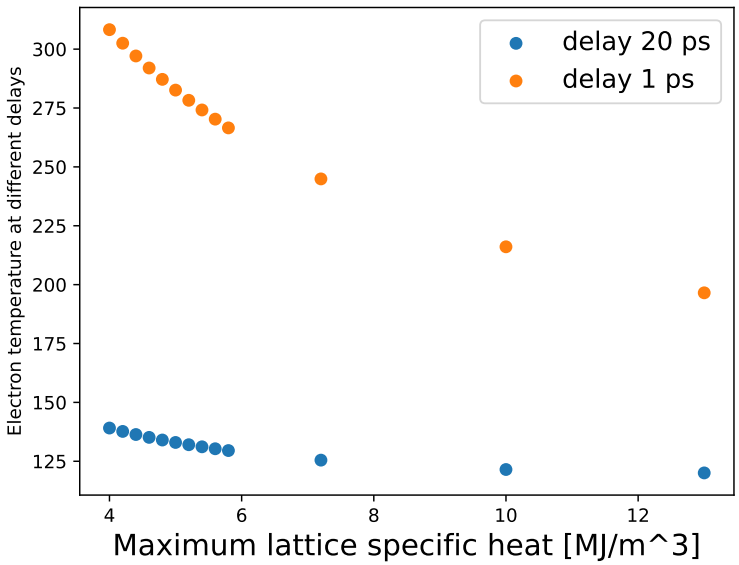
Magnetization and electron tempereature in multi-lattice approach



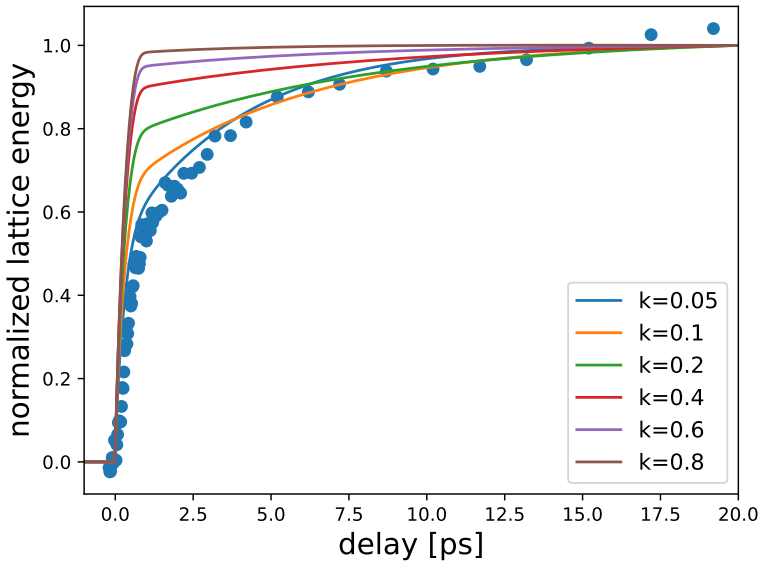


Lattice heat capacity

Maximum lattice heat capacity for $k = 0.1$



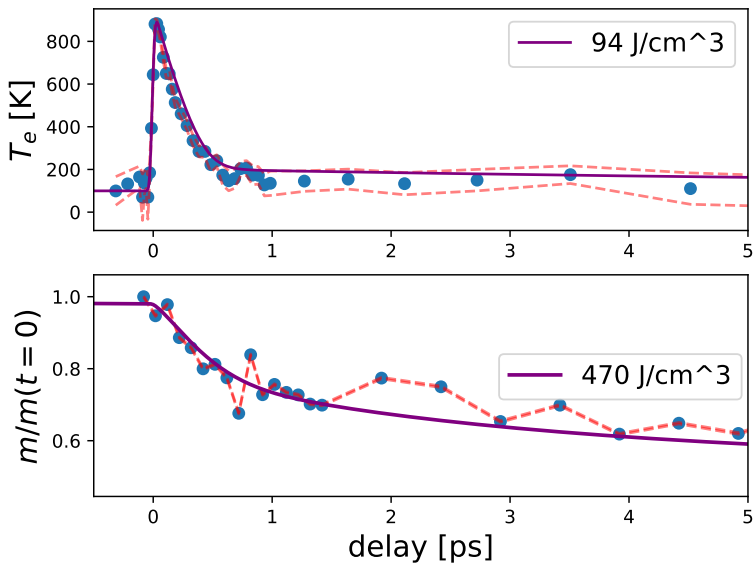
Distribution of lattice heat capacity



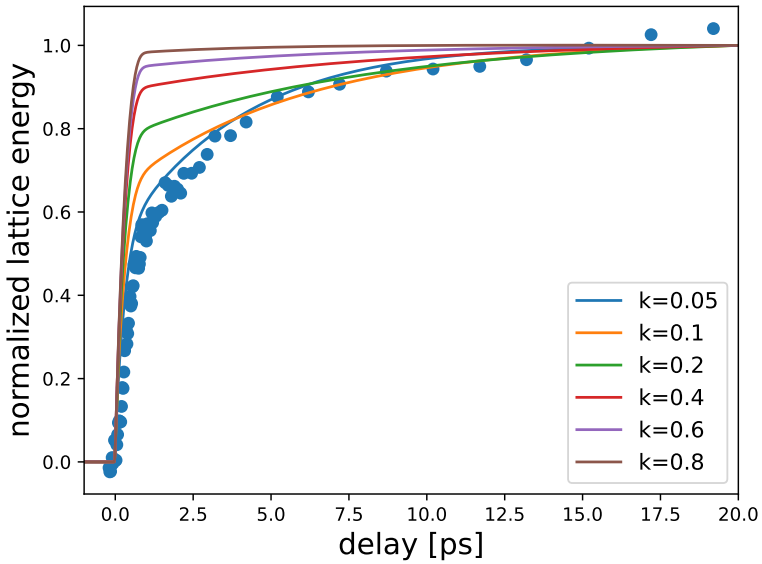
$$C_{po} = kC_p(T_{po})T_{po}$$

Lattice heat capacity

Electron and spin systems



Distribution of lattice heat capacity



$$C_{po} = kC_p(T_{po})T_{po}$$