The role of the magnon gap in spin-charge conversion at magnetic insulator/heavy metal interfaces

Joseph Barker

j.barker@leeds.ac.uk

University of Leeds

Engineering and Physical Sciences Research Council

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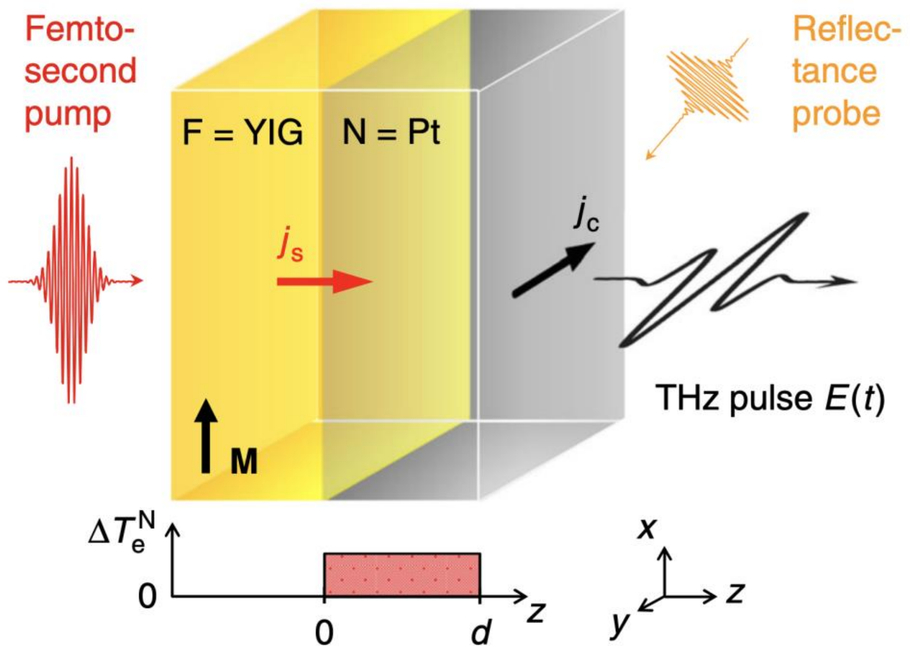
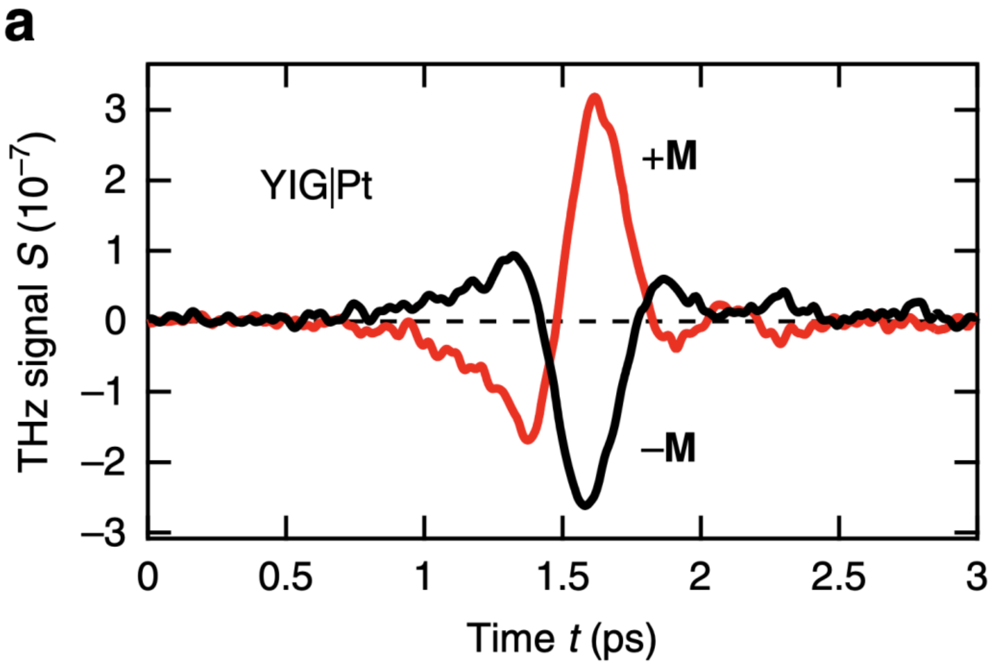
THz emission from insulator/metal multilayers

Femtosecond formation dynamics of the spin Seebeck effect revealed by terahertz spectroscopy



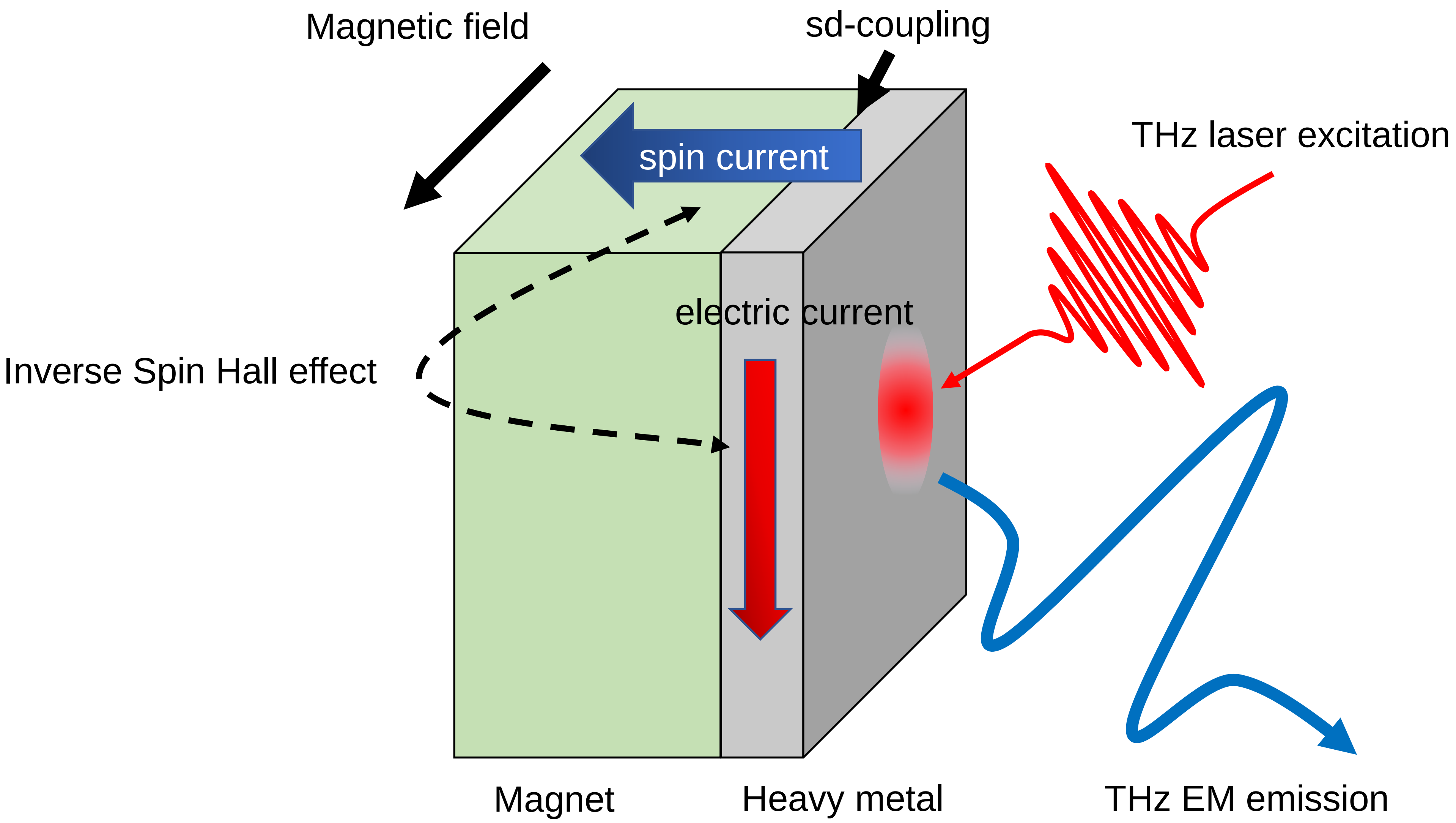


T.S. Seifert *et al.*, Nat. Commun. **9**, 2899 (2018)

Not the ‘usual’ spin Seebeck effect

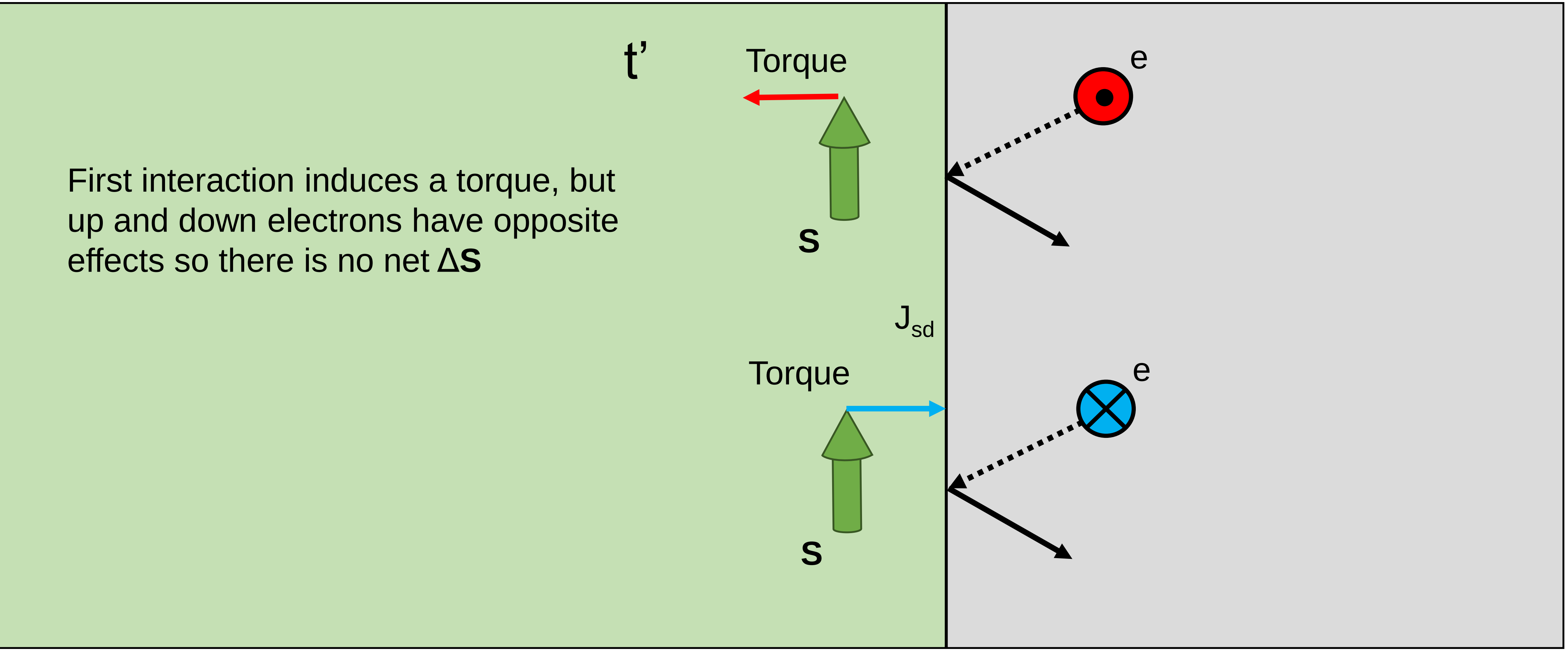
- Too fast for temperature gradients to be established in the magnet.
- Only the metal is heated.
- THz experiments remove the long ranged spin transport of a temperature gradient - sensitive only to the interface spin-charge conversion.

Overview


Magnetic field
sd-coupling
THz laser excitation
spin current
electric current
Inverse Spin Hall effect
Magnet
Heavy metal
THz EM emission

Interface spin currents - spin transfer torques

The electron spin polarisation is **perpendicular** to the magnetic moments

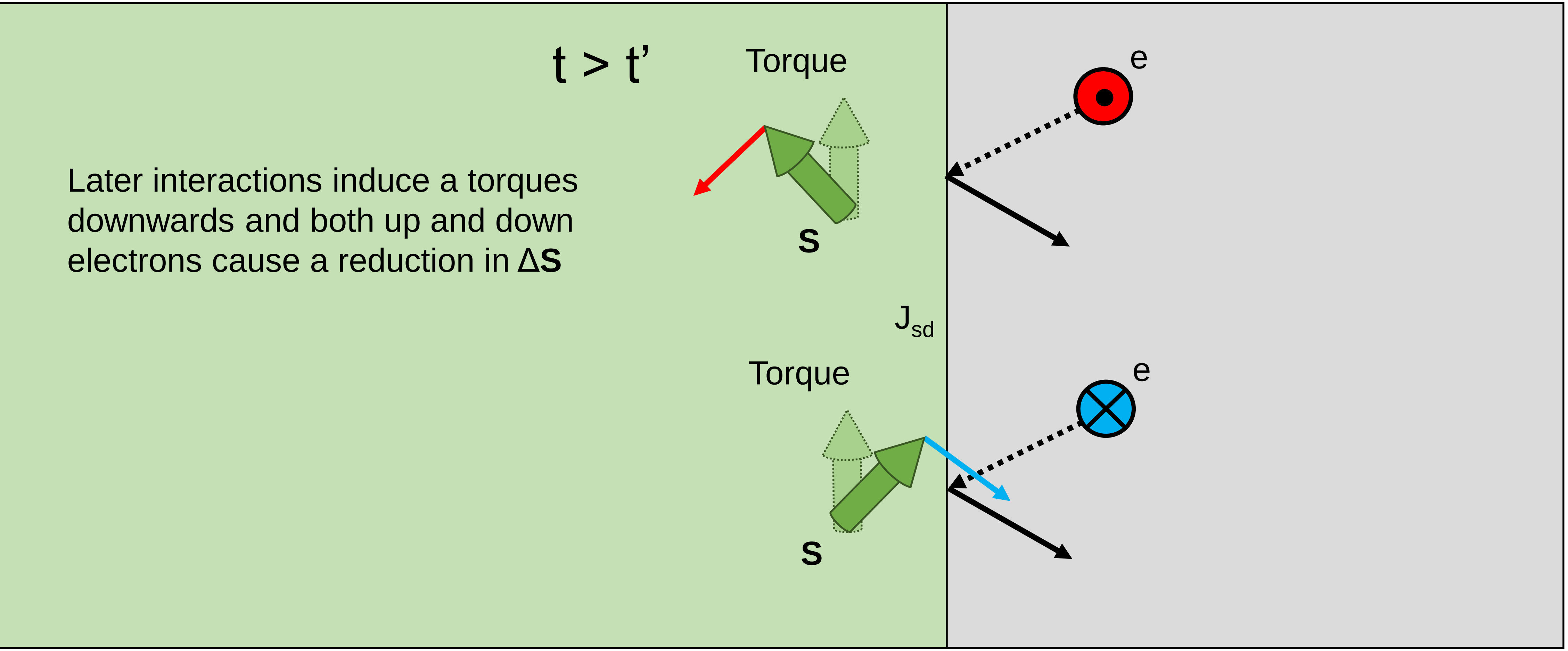


First interaction induces a torque, but up and down electrons have opposite effects so there is no net Δ**S**

T.S. Seifert *et al.*, Nat. Commun. **9**, 2899 (2018)

Interface spin currents - spin transfer torques

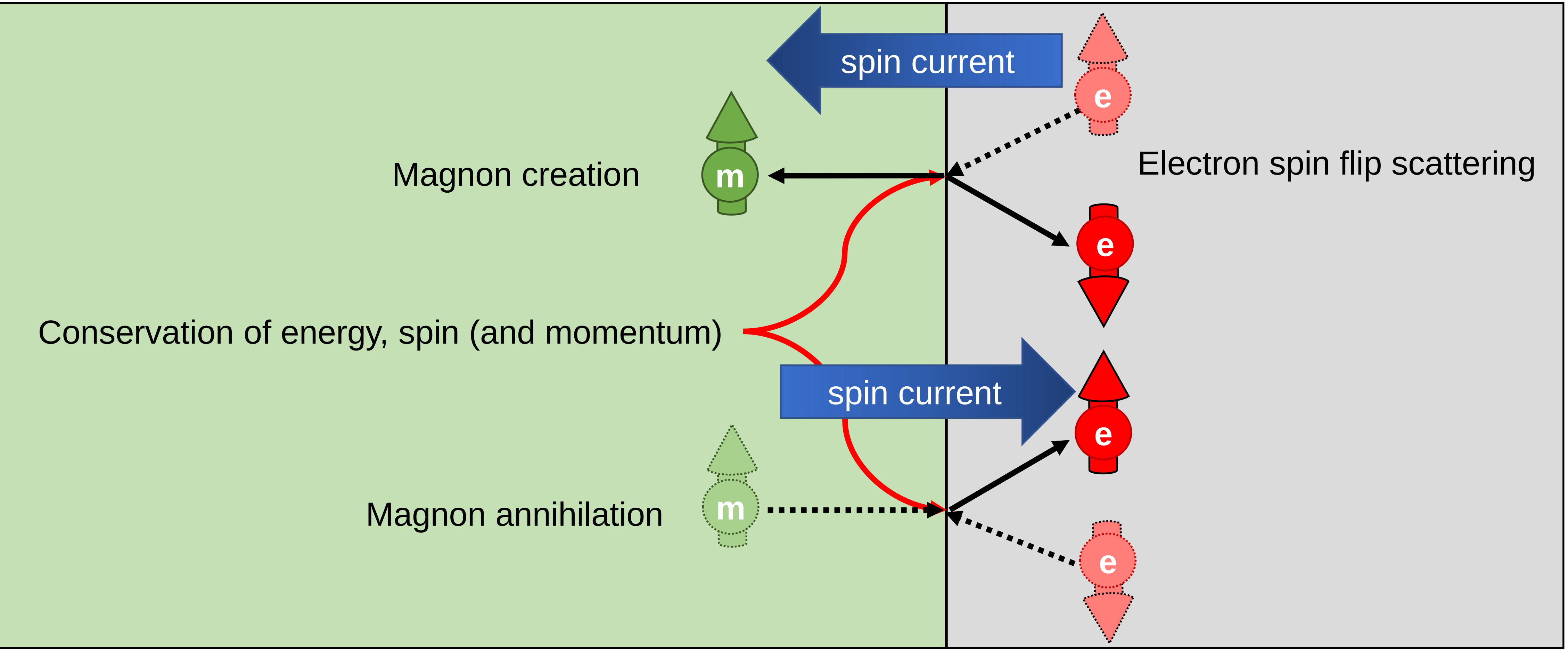
The electron spin polarisation is **perpendicular** to the magnetic moments



T.S. Seifert *et al.*, Nat. Commun. **9**, 2899 (2018)

Interface spin currents – incoherent scattering

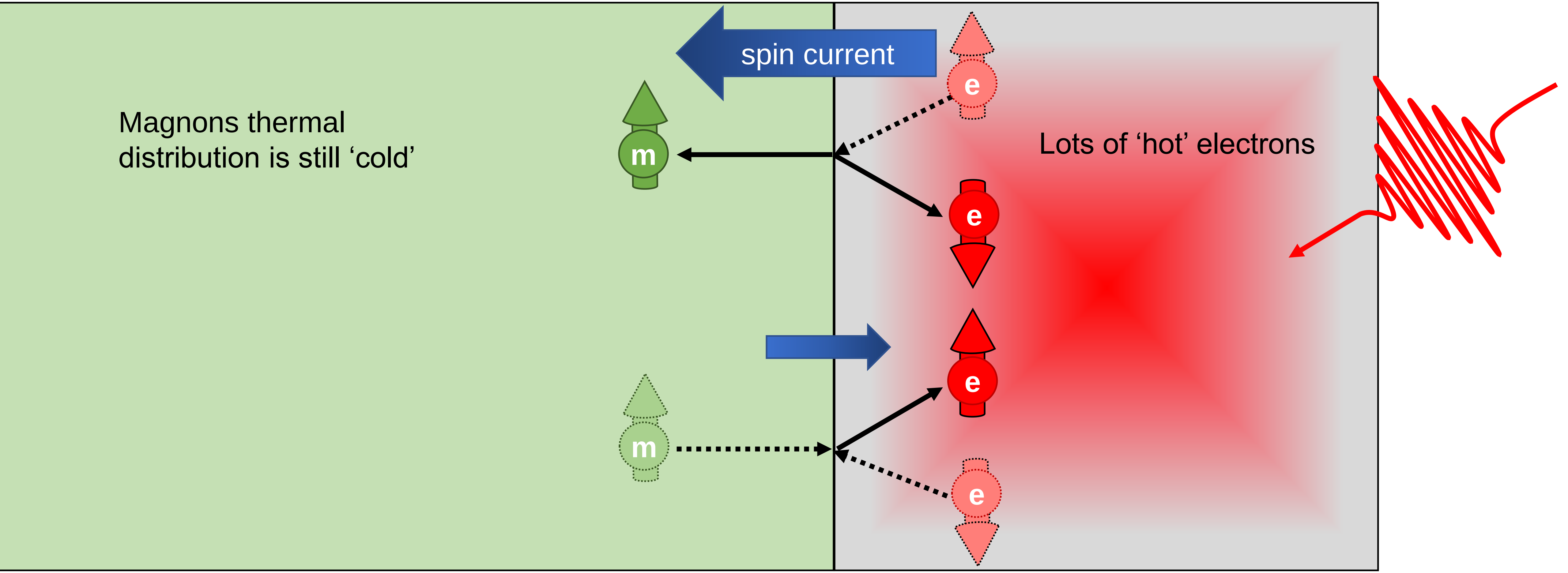
The electron spin polarisation is **parallel** to the magnetic moments



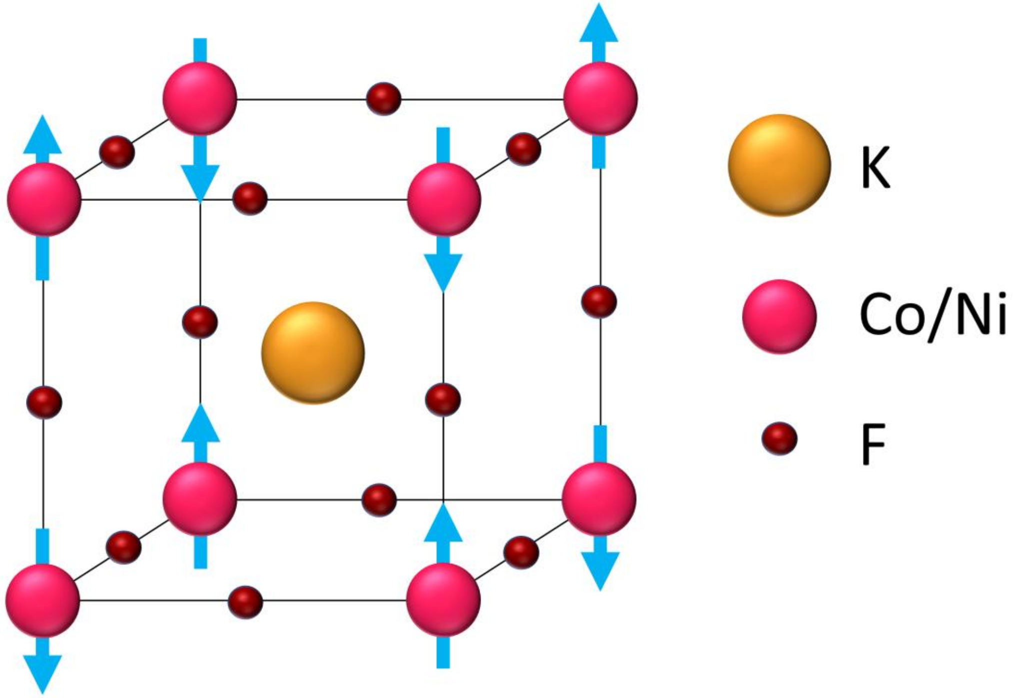
S.A. Bender & Y. Tserkovnyak, Phys. Rev. B **91**, 140402 (2015)

Interface spin currents

Ultrafast heating of the heavy metal electrons breaking the equilibrium and leading to a net spin current



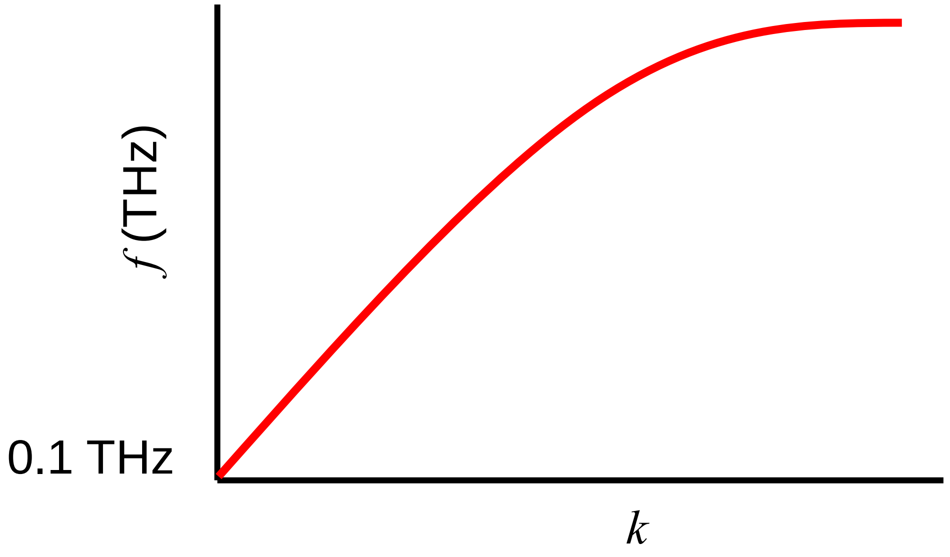
$KCoF_3$ and $KNiF_3$



- Antiferromagnetic insulators
- Identical structure and magnetic space group
- Different magnetic anisotropy

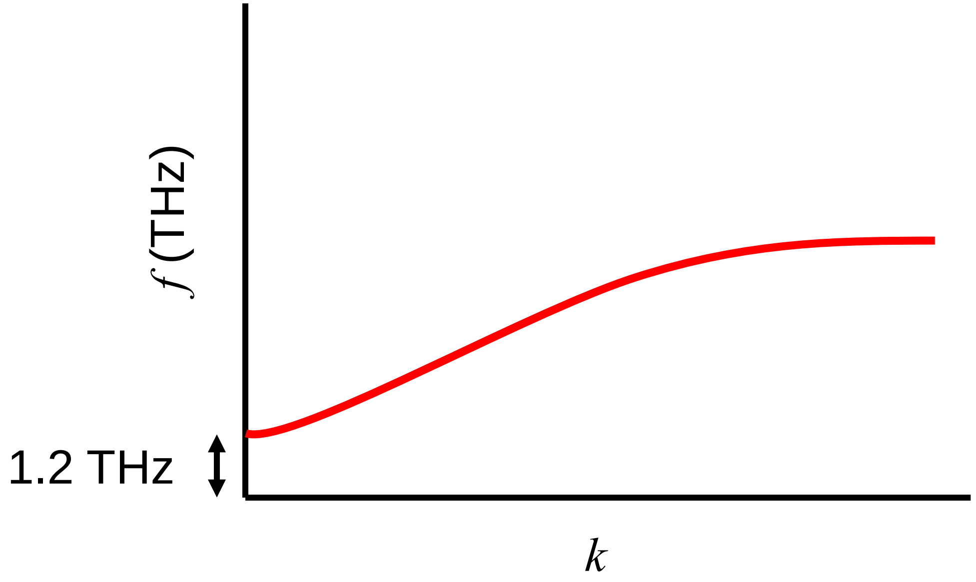
Magnon dispersion

$KNiF_3$



Similar order of magnitude to ferromagnets

$KCoF_3$

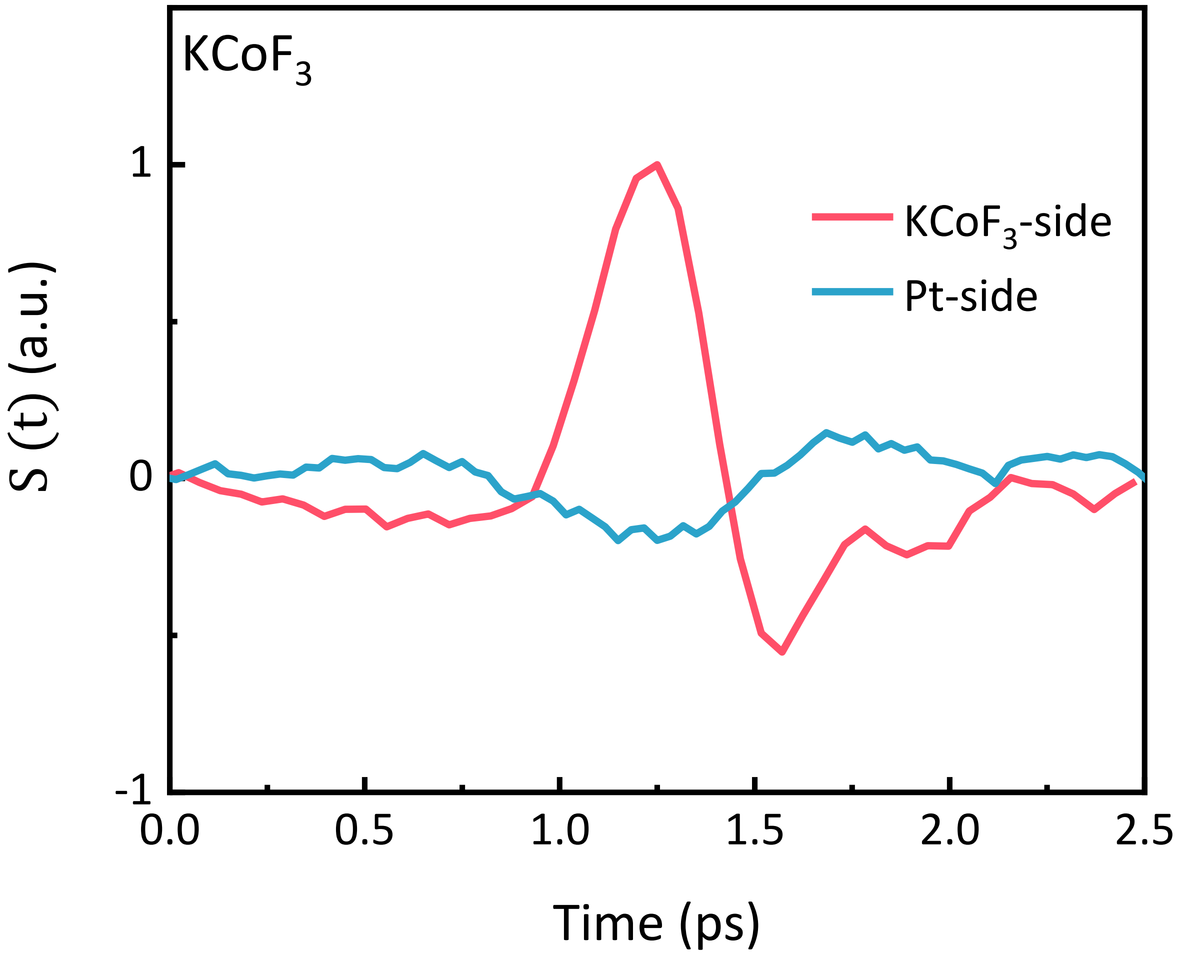


1-2 Orders of magnitude larger than ferromagnets

The gap probably plays a role, but how do we interpret this for these very complex non-equilibrium experiments.

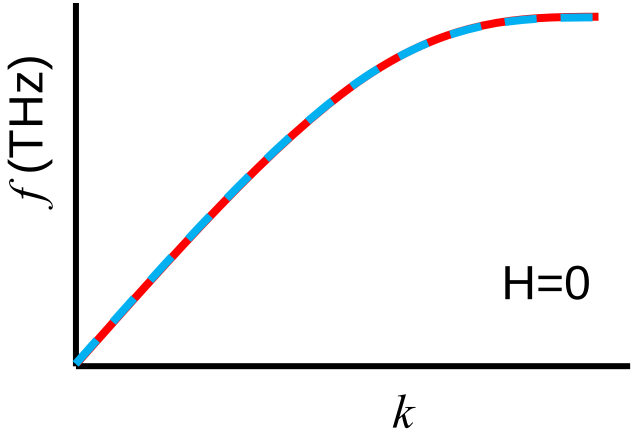
THz signal

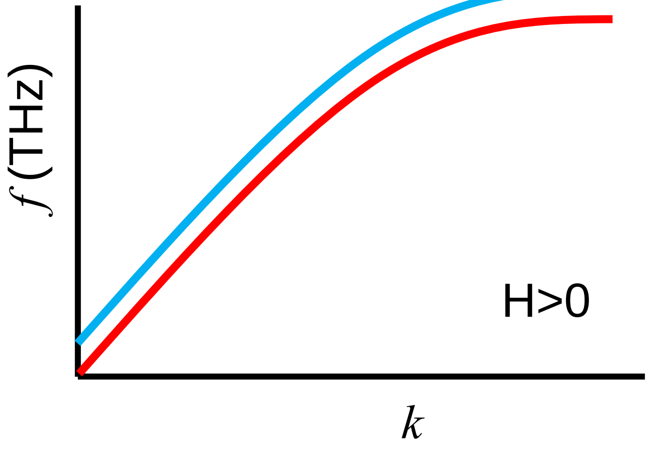
Emission only lasts for 2 picoseconds.

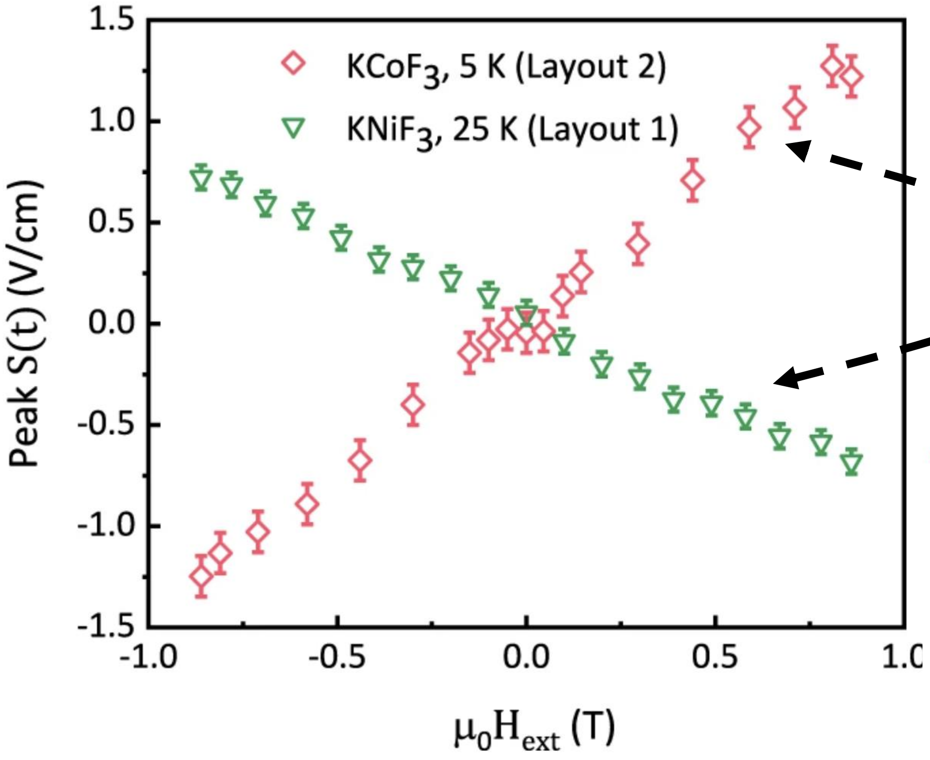


F.N. Kholid *et al.*, Nat. Commun. **14**, 538 (2023)

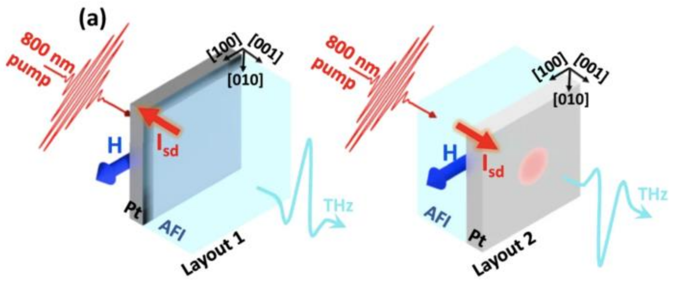
THz signal is field dependent







Opposite dependence because of different geometries

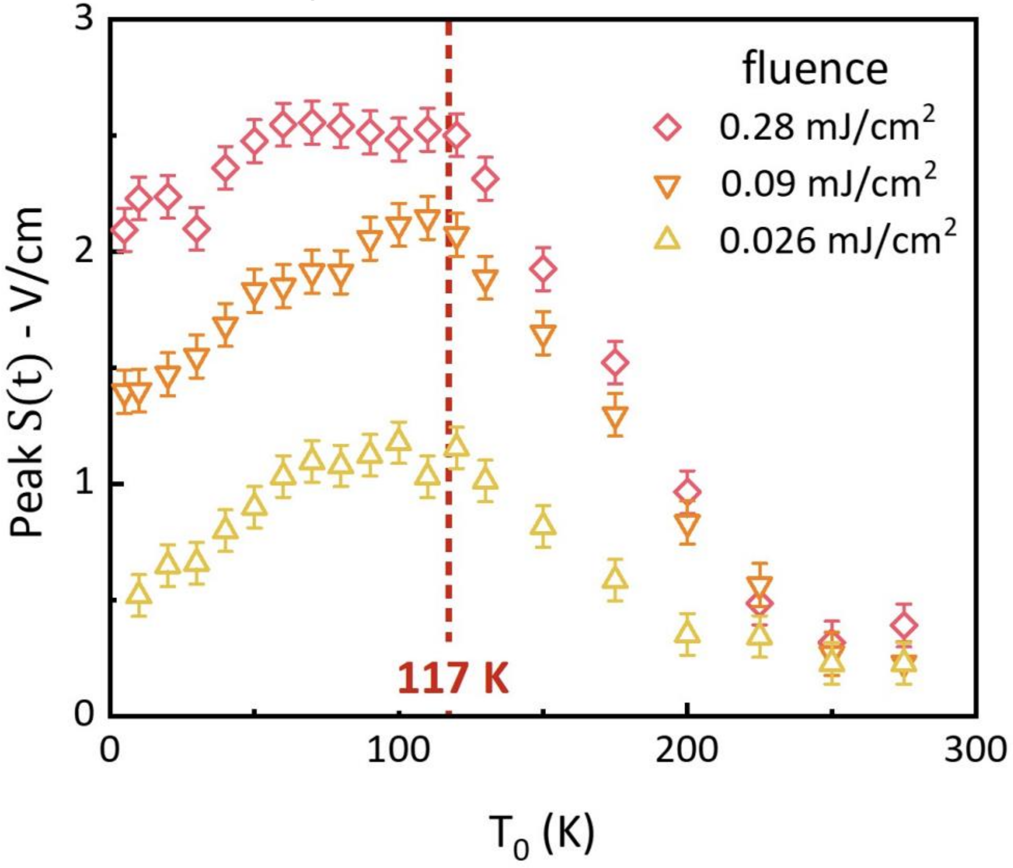


Two degenerate modes that must be split with a field to generate a net spin current

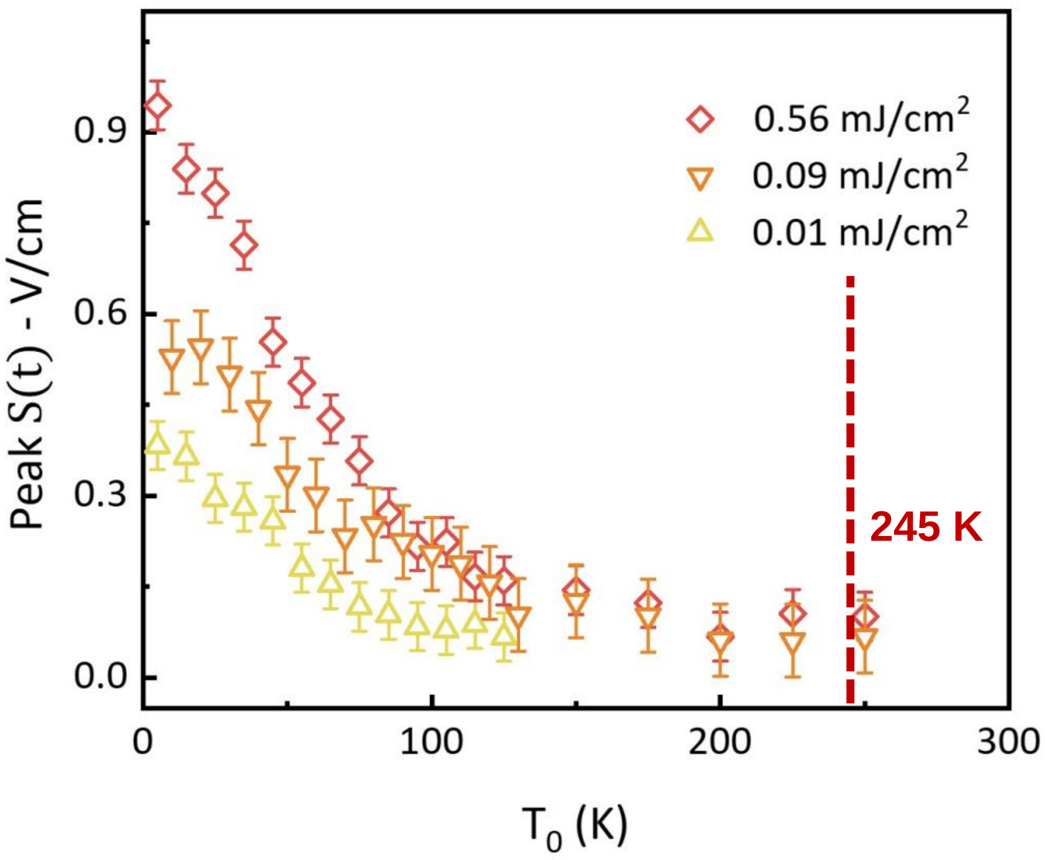
F.N. Kholid *et al.*, Nat. Commun. **14**, 538 (2023)

Temperature dependence

$KCoF_3/Pt$



$KNiF_3/Pt$

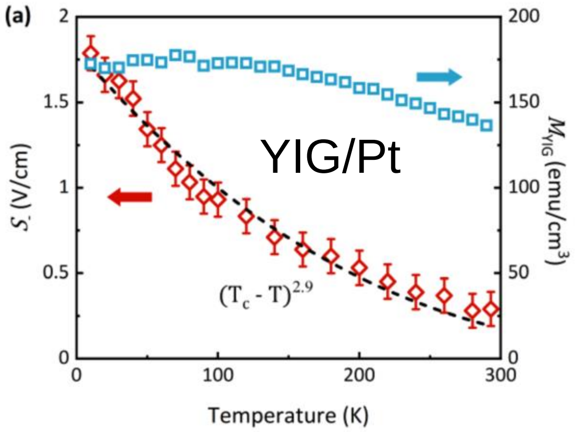


F.N. Kholid *et al.*, Nat. Commun. **14**, 538 (2023)

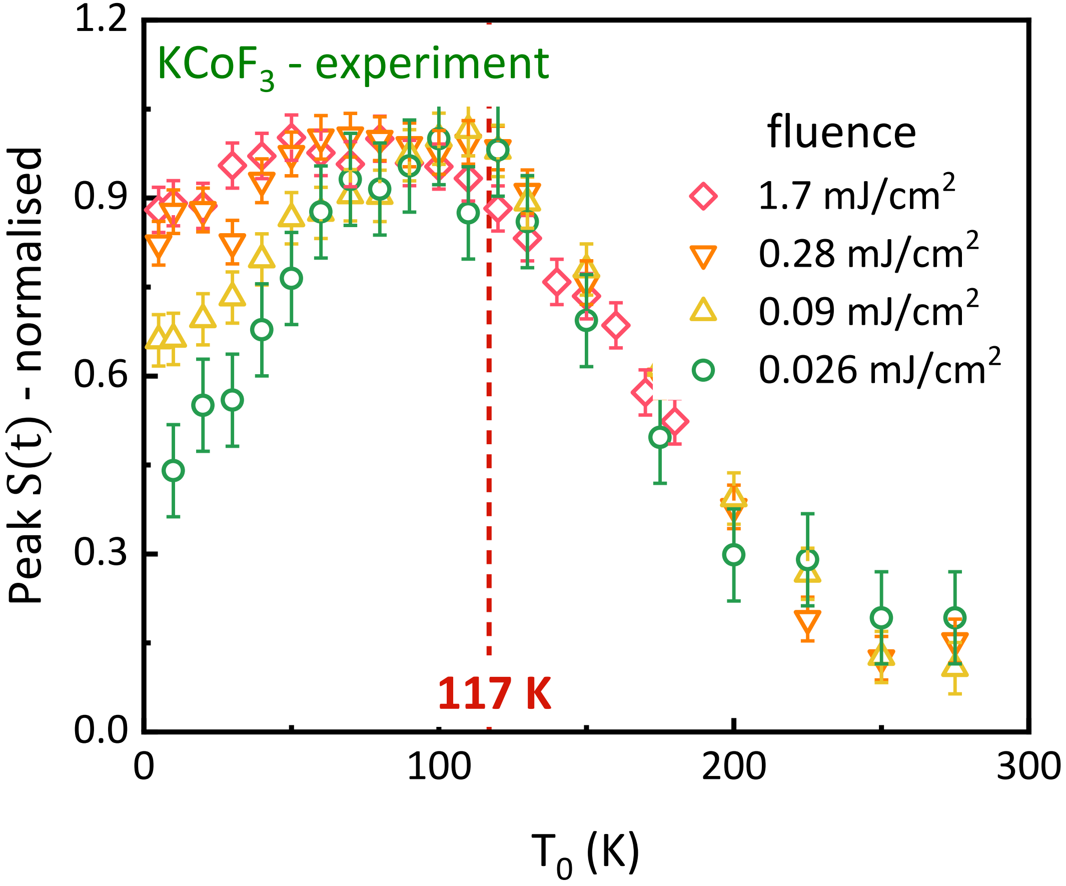
THz signal amplitude

F.N. Kholid *et al.*, Appl. Phys. Lett. **119**, 032401 (2021)

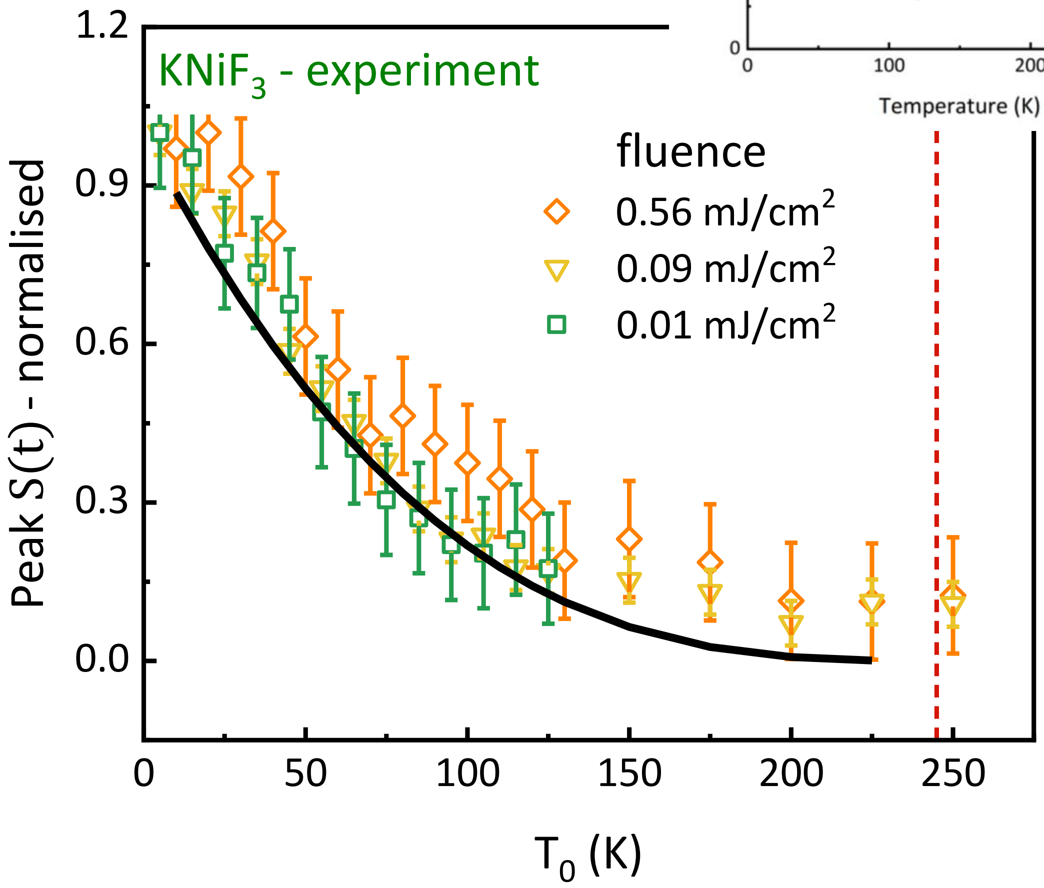
Temperature dependence of spin mixing conductance



$KCoF_3$/Pt



$KNiF_3$/Pt



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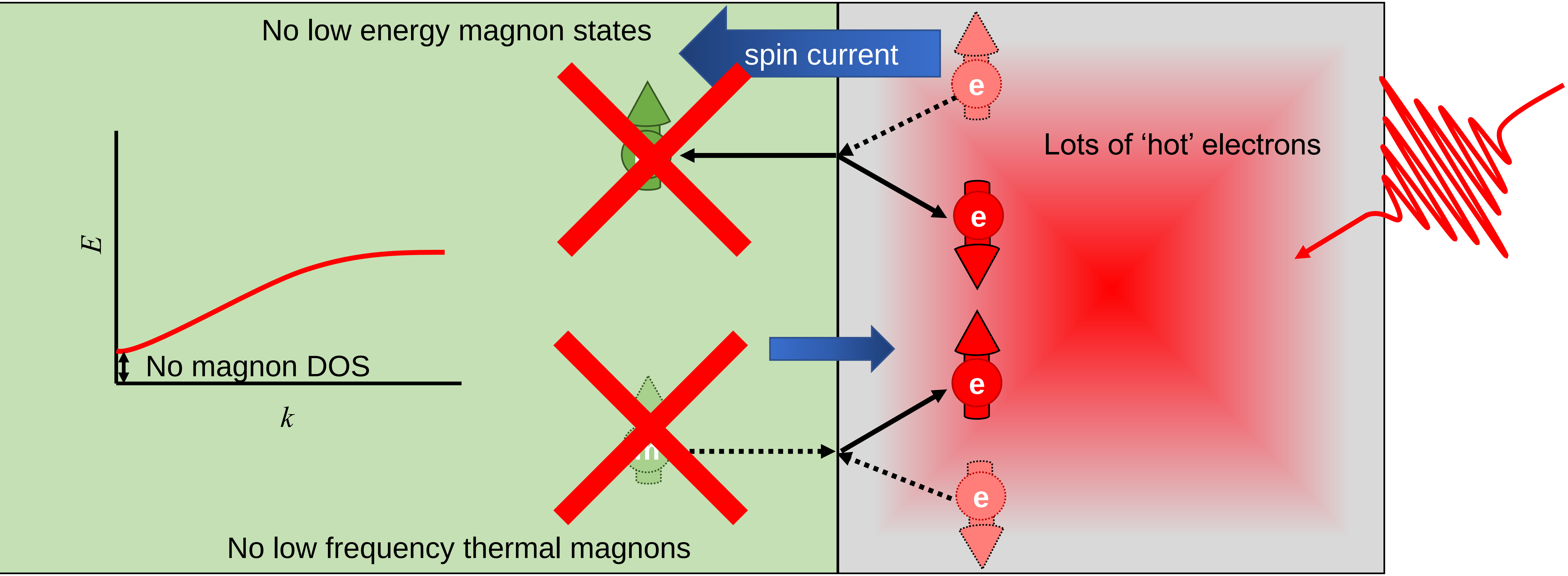
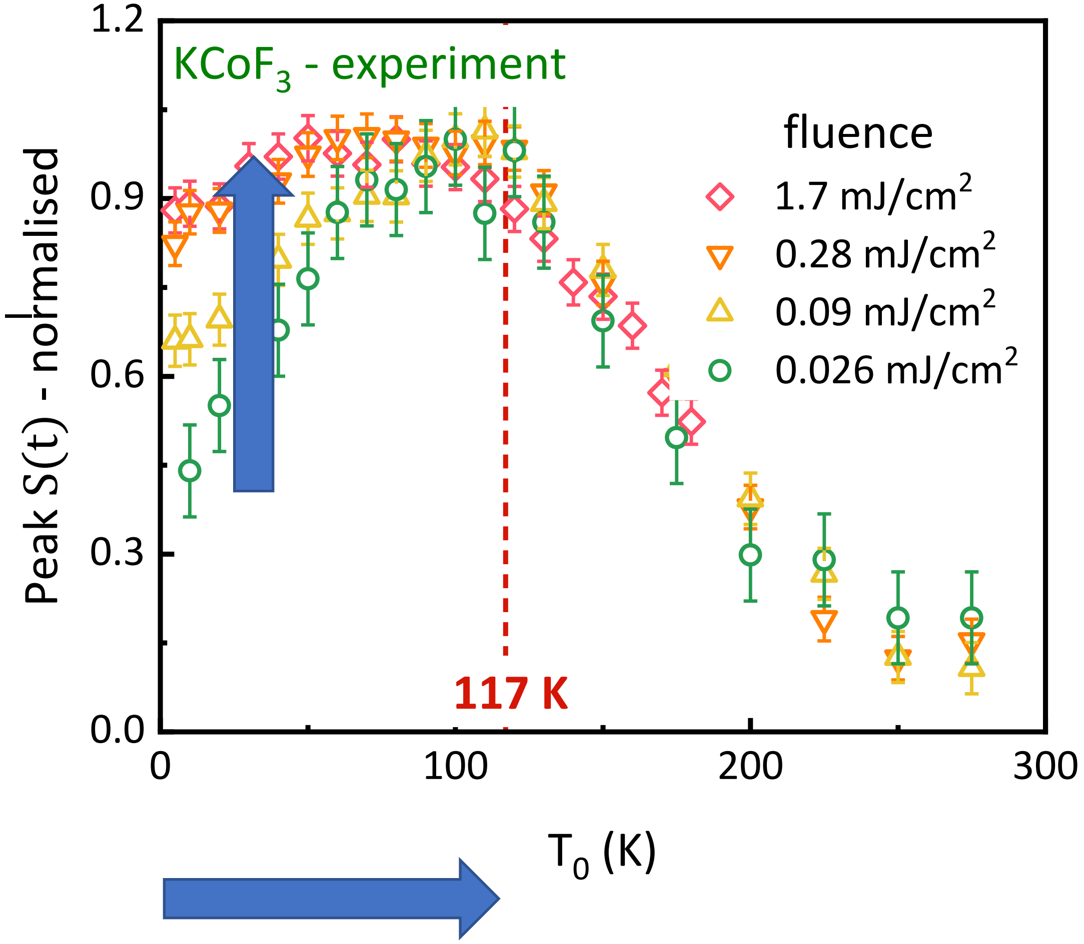
Interface spin currents – mDOS gap

If there are no magnon states, then electrons with energy below the gap cannot transfer spin



$KCoF_3$



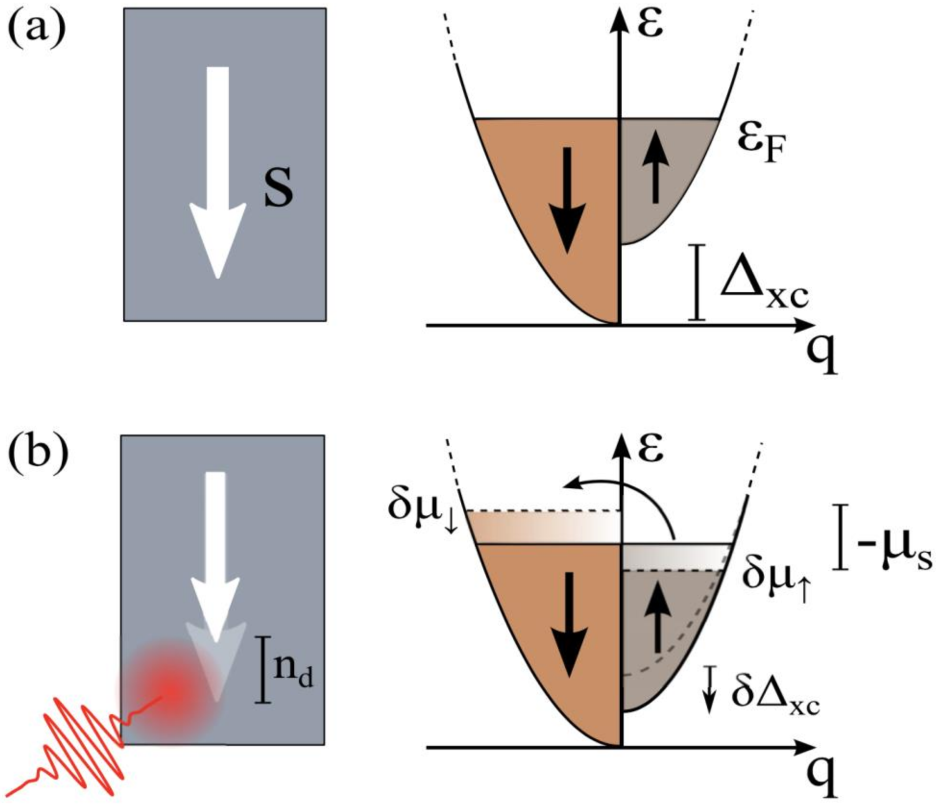
Higher laser fluence produces higher energy electrons – overcomes the gap

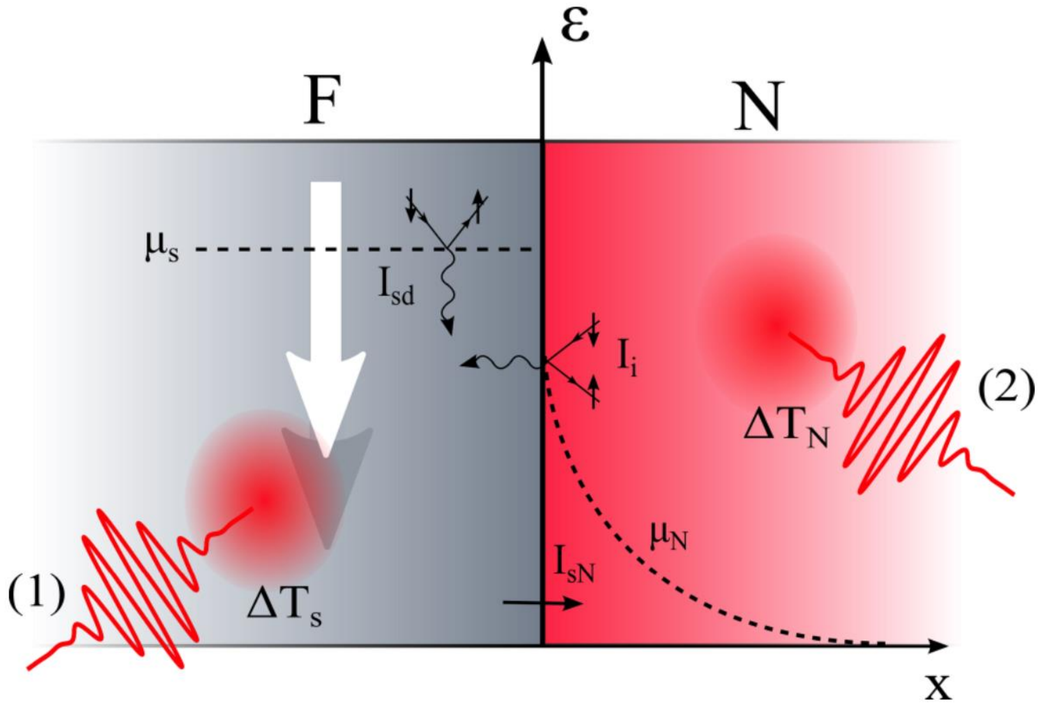
Hotter ambient temperature means more thermal electrons above the gap

F.N. Kholid *et al.*, Nat. Commun. **14**, 538 (2023)

Electron-magnon scattering in magnetic heterostructures far out of equilibrium

Tveten, Phys. Rev. B **92**, 180412 (2015)





"Just" need to derive for an antiferromagnetic insulator!

Theoretical model

$$\mathscr{H}_{\mathrm{AFM}} = J_{\mathrm{AFM}} \sum_{i,j} \mathbf{S}_j \cdot \mathbf{S}_j + H \sum_i S_i^x - K \sum_i (S_i^x)^2$$

Heisenberg model for antiferromagnet

$$\mathscr{H}_{\mathrm{Pt}} = \sum_{i,j} \left(t_{ij} c_i^\dagger c_j + \mathrm{h.c.} \right)$$

Tight binding model for metal

$$\mathscr{H}_{sd} = \sum_i J_{sd} \mathbf{S}_i \cdot c_i^\dagger \boldsymbol{\sigma} c_i$$

sd coupling at the interface

- Assume electrons follow a thermal (Fermi-Dirac) distribution
- Magnon distribution is allowed to be non-equilibrium
- Apply Fermi's Golden Rule for the *sd* scattering

Do a little maths...

$$
\begin{aligned}
I_{\mathrm{sd}} = \frac{\pi S J^2}{A} \sum_{\mathbf{k}} {\sum_{\mathbf{q}}}' \Bigg[& \xi_{\mathbf{q}}^2 [1 + n_\alpha(\varepsilon_{-\mathbf{q}}^\alpha)] n_B(\varepsilon_{\mathbf{k}+\mathbf{q}} - \varepsilon_{\mathbf{k}} - \mu_\downarrow + \mu_\uparrow)[n_F(\varepsilon_{\mathbf{k}} - \mu_\uparrow) - n_F(\varepsilon_{\mathbf{k}+\mathbf{q}} - \mu_\downarrow)] \delta(\varepsilon_{\mathbf{k}} + \varepsilon_{-\mathbf{q}}^\alpha - \varepsilon_{\mathbf{k}+\mathbf{q}}) \\
& - \xi_{\mathbf{q}}^2 [n_\alpha(\varepsilon_{-\mathbf{q}}^\alpha)] n_B(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}} + \mu_\downarrow - \mu_\uparrow)[n_F(\varepsilon_{\mathbf{k}+\mathbf{q}} - \mu_\downarrow) - n_F(\varepsilon_{\mathbf{k}} - \mu_\uparrow)] \delta(\varepsilon_{\mathbf{k}} + \varepsilon_{-\mathbf{q}}^\alpha - \varepsilon_{\mathbf{k}+\mathbf{q}}) \\
& + \xi_{\mathbf{q}}^2 [n_\beta(\varepsilon_{\mathbf{q}}^\beta)] n_B(\varepsilon_{\mathbf{k}+\mathbf{q}} - \varepsilon_{\mathbf{k}} - \mu_\downarrow + \mu_\uparrow)[n_F(\varepsilon_{\mathbf{k}} - \mu_\uparrow) - n_F(\varepsilon_{\mathbf{k}+\mathbf{q}} - \mu_\downarrow)] \delta(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{q}}^\beta - \varepsilon_{\mathbf{k}+\mathbf{q}}) \\
& - \xi_{\mathbf{q}}^2 [1 + n_\beta(\varepsilon_{\mathbf{q}}^\beta)] n_B(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}} + \mu_\downarrow - \mu_\uparrow)[n_F(\varepsilon_{\mathbf{k}+\mathbf{q}} - \mu_\downarrow) - n_F(\varepsilon_{\mathbf{k}} - \mu_\uparrow)] \delta(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{q}}^\beta - \varepsilon_{\mathbf{k}+\mathbf{q}}) \\
& + \xi_{\mathbf{q}}^{-2} [1 + n_\alpha(\varepsilon_{-\mathbf{q}}^\alpha)] n_B(\varepsilon_{\mathbf{k}+\mathbf{q}+\mathbf{Q}} - \varepsilon_{\mathbf{k}} - \mu_\downarrow + \mu_\uparrow)[n_F(\varepsilon_{\mathbf{k}} - \mu_\uparrow) - n_F(\varepsilon_{\mathbf{k}+\mathbf{q}+\mathbf{Q}} - \mu_\downarrow)] \delta(\varepsilon_{\mathbf{k}} + \varepsilon_{-\mathbf{q}}^\alpha - \varepsilon_{\mathbf{k}+\mathbf{q}+\mathbf{Q}}) \\
& - \xi_{\mathbf{q}}^{-2} [n_\alpha(\varepsilon_{-\mathbf{q}}^\alpha)] n_B(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}+\mathbf{Q}} + \mu_\downarrow - \mu_\uparrow)[n_F(\varepsilon_{\mathbf{k}+\mathbf{q}+\mathbf{Q}} - \mu_\downarrow) - n_F(\varepsilon_{\mathbf{k}} - \mu_\uparrow)] \delta(\varepsilon_{\mathbf{k}} + \varepsilon_{-\mathbf{q}}^\alpha - \varepsilon_{\mathbf{k}+\mathbf{q}+\mathbf{Q}}) \\
& + \xi_{\mathbf{q}}^{-2} [n_\beta(\varepsilon_{\mathbf{q}}^\beta)] n_B(\varepsilon_{\mathbf{k}+\mathbf{q}+\mathbf{Q}} - \varepsilon_{\mathbf{k}} - \mu_\downarrow + \mu_\uparrow)[n_F(\varepsilon_{\mathbf{k}} - \mu_\uparrow) - n_F(\varepsilon_{\mathbf{k}+\mathbf{q}+\mathbf{Q}} - \mu_\downarrow)] \delta(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{q}}^\beta - \varepsilon_{\mathbf{k}+\mathbf{q}+\mathbf{Q}}) \\
& - \xi_{\mathbf{q}}^{-2} [1 + n_\beta(\varepsilon_{\mathbf{q}}^\beta)] n_B(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}+\mathbf{Q}} + \mu_\downarrow - \mu_\uparrow)[n_F(\varepsilon_{\mathbf{k}+\mathbf{q}+\mathbf{Q}} - \mu_\downarrow) - n_F(\varepsilon_{\mathbf{k}} - \mu_\uparrow)] \delta(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{q}}^\beta - \varepsilon_{\mathbf{k}+\mathbf{q}+\mathbf{Q}}) \Bigg]
\end{aligned}
$$

Dynamical description

Magnon dispersion

$$\varepsilon_k \approx 4\sqrt{3}SJ_{\mathrm{AFM}}k + 2S\sqrt{(J_{\mathrm{AFM}}z + K)K} \mp g\mu_B B$$

Instantaneous spin current

$$I_{\mathrm{sd}} = \sqrt{\frac{S}{2}}\left\{ \int_{\varepsilon_0^\alpha}^{\varepsilon_{\max}^\alpha} d\varepsilon_q^\alpha \Gamma(\varepsilon_q^\alpha) g_\alpha(\varepsilon_q^\alpha) \left[(u_q + v_q)^2 + (u_q - v_q)^2\right] [\varepsilon_q^\alpha + \mu_s][n_B(\varepsilon_q^\alpha + \mu_s) - n_\alpha(\varepsilon_q^\alpha)] \right.$$
$$\left. - \int_{\varepsilon_0^\beta}^{\varepsilon_{\max}^\beta} d\varepsilon_q^\beta \Gamma(\varepsilon_q^\beta) g_\beta(\varepsilon_q^\beta) \left[(u_q + v_q)^2 + (u_q - v_q)^2\right] [\varepsilon_q^\beta - \mu_s][n_B(\varepsilon_q^\beta - \mu_s) - n_\beta(\varepsilon_q^\beta)] \right\}$$

Electron temperature dynamics
(induced by the laser heating)

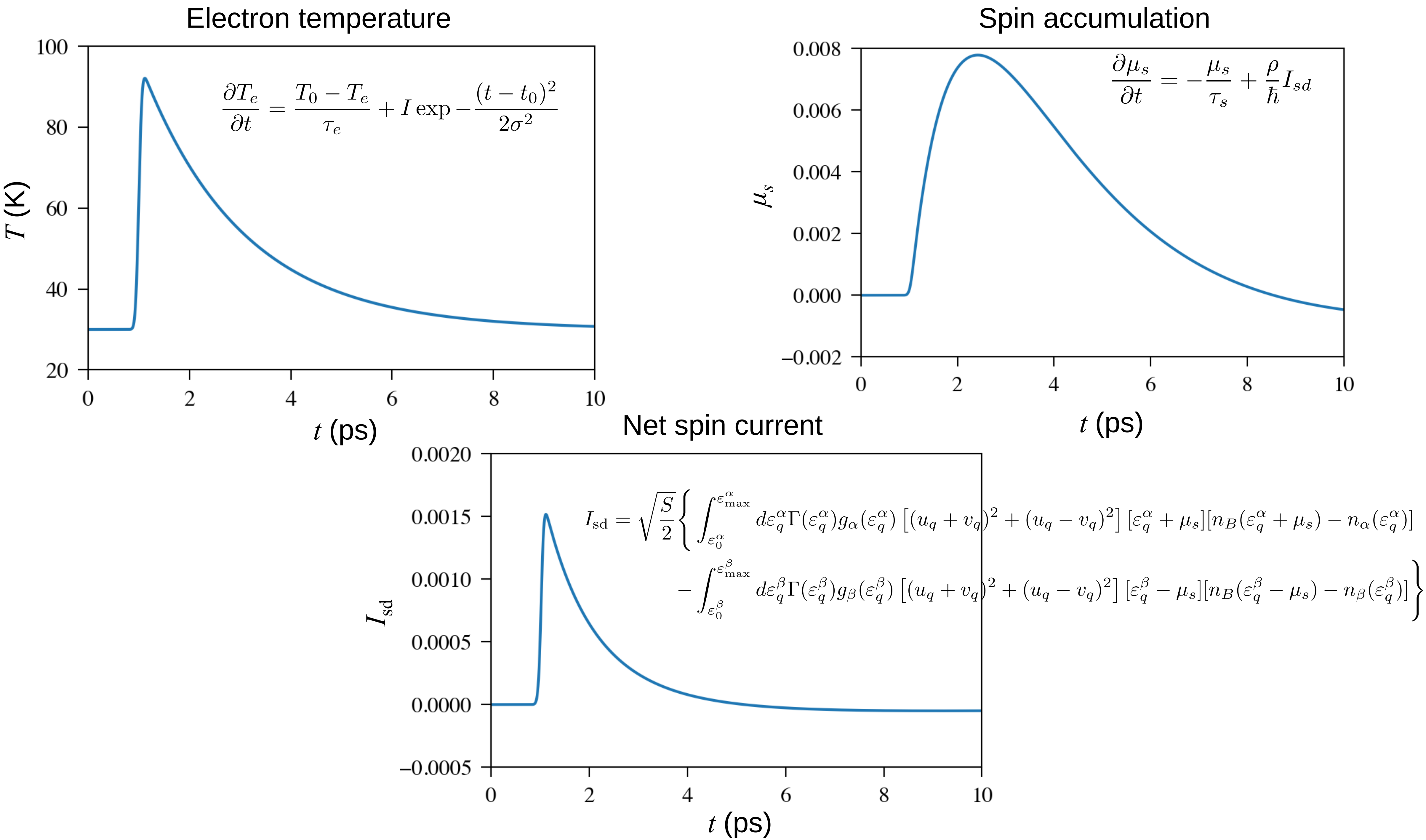
$$\frac{\partial T_e}{\partial t} = \frac{T_0 - T_e}{\tau_e} + I\exp - \frac{(t - t_0)^2}{2\sigma^2}$$

Spin accumulation dynamics
(induced by the non-equilibrium spin current)

$$\frac{\partial \mu_s}{\partial t} = -\frac{\mu_s}{\tau_s} + \frac{\rho}{\hbar} I_{sd}$$

Magnon population dynamics

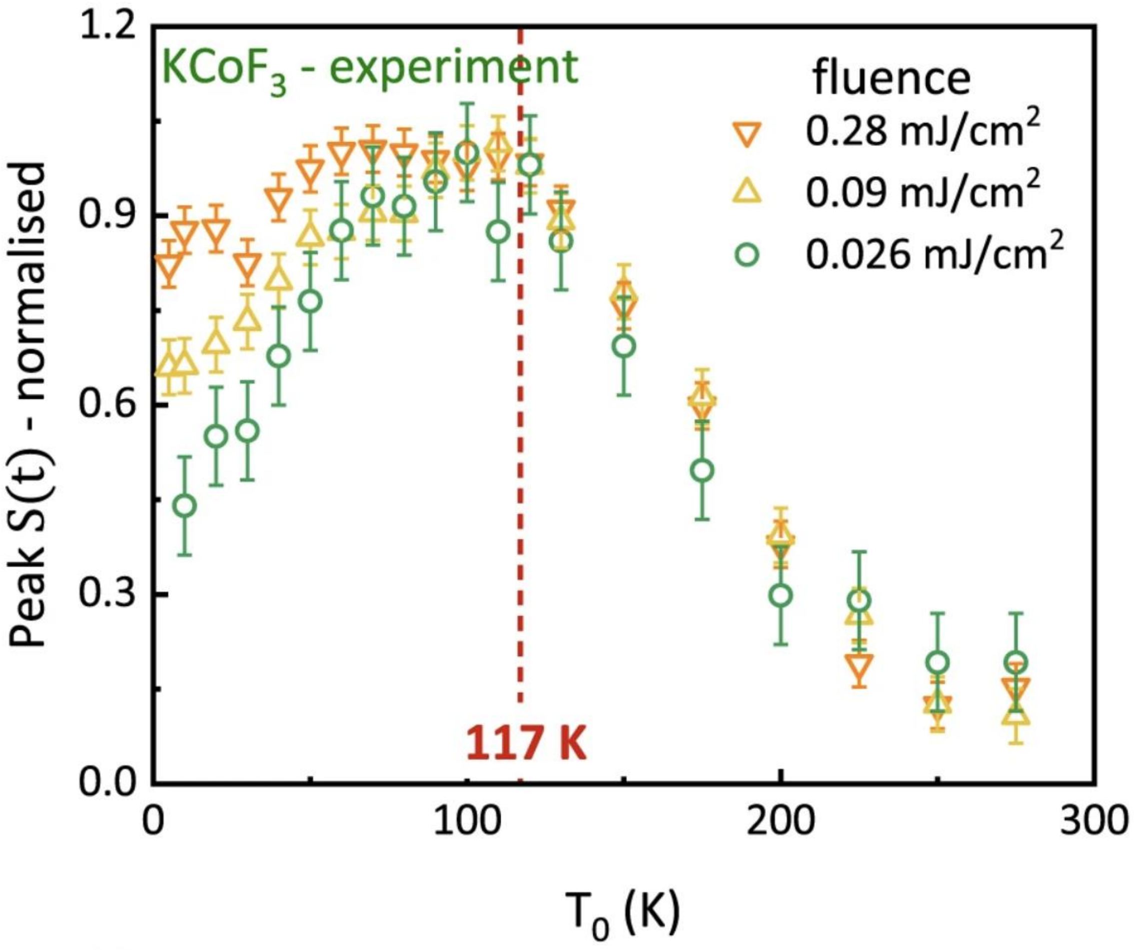
$$\frac{\partial n_\alpha(\varepsilon_q^\alpha)}{\partial t} = \frac{I_{sd}(\varepsilon_q^\alpha)}{\hbar} = \frac{1}{\hbar}\sqrt{\frac{S}{2}}\Gamma(\varepsilon_q^\alpha) g_\alpha(\varepsilon_q^\alpha) \left[(u_q + v_q)^2 + (u_q - v_q)^2\right] [\varepsilon_q^\alpha + \mu_s][n_B(\varepsilon_q^\alpha + \mu_s) - n_\alpha(\varepsilon_q^\alpha)]$$



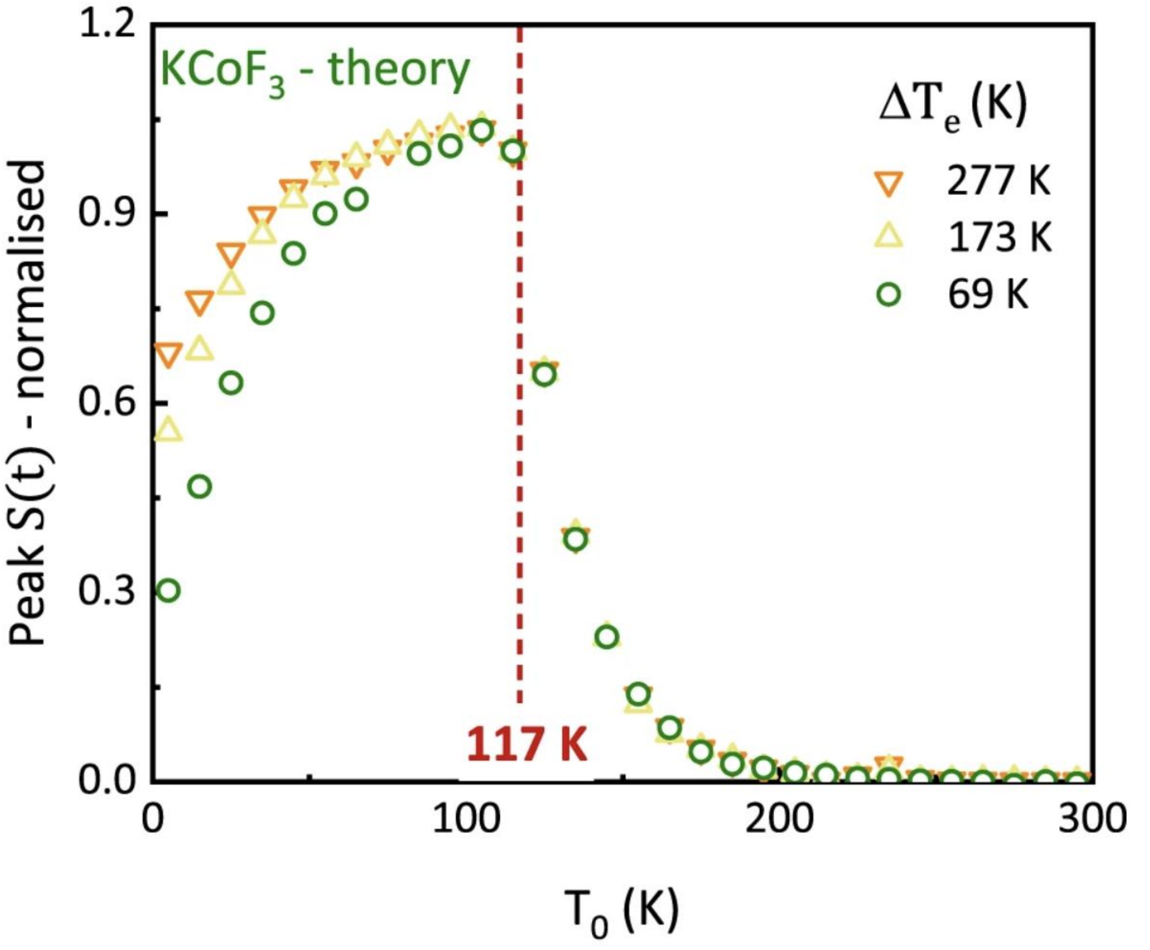
Electron temperature
$\frac{\partial T_e}{\partial t} = \frac{T_0 - T_e}{\tau_e} + I \exp - \frac{(t-t_0)^2}{2\sigma^2}$
T (K)
100
80
60
40
20
0
2
4
6
8
10
t (ps)
Spin accumulation
$\frac{\partial \mu_s}{\partial t} = -\frac{\mu_s}{\tau_s} + \frac{\rho}{\hbar} I_{sd}$
μ_s
0.008
0.006
0.004
0.002
0.000
−0.002
0
2
4
6
8
10
t (ps)
Net spin current
$I_{\rm sd} = \sqrt{\frac{S}{2}} \Big\{ \int_{\varepsilon_0^\alpha}^{\varepsilon_{\rm max}^\alpha} d\varepsilon_q^\alpha \Gamma(\varepsilon_q^\alpha) g_\alpha(\varepsilon_q^\alpha) \left[(u_q+v_q)^2 + (u_q-v_q)^2\right] [\varepsilon_q^\alpha + \mu_s][n_B(\varepsilon_q^\alpha + \mu_s) - n_\alpha(\varepsilon_q^\alpha)]$
$- \int_{\varepsilon_0^\beta}^{\varepsilon_{\rm max}^\beta} d\varepsilon_q^\beta \Gamma(\varepsilon_q^\beta) g_\beta(\varepsilon_q^\beta) \left[(u_q+v_q)^2 + (u_q-v_q)^2\right] [\varepsilon_q^\beta - \mu_s][n_B(\varepsilon_q^\beta - \mu_s) - n_\beta(\varepsilon_q^\beta)] \Big\}$
$I_{\rm sd}$
0.0020
0.0015
0.0010
0.0005
0.0000
−0.0005
0
2
4
6
8
10
t (ps)

Comparing theory and experiments

Experiment



Theory



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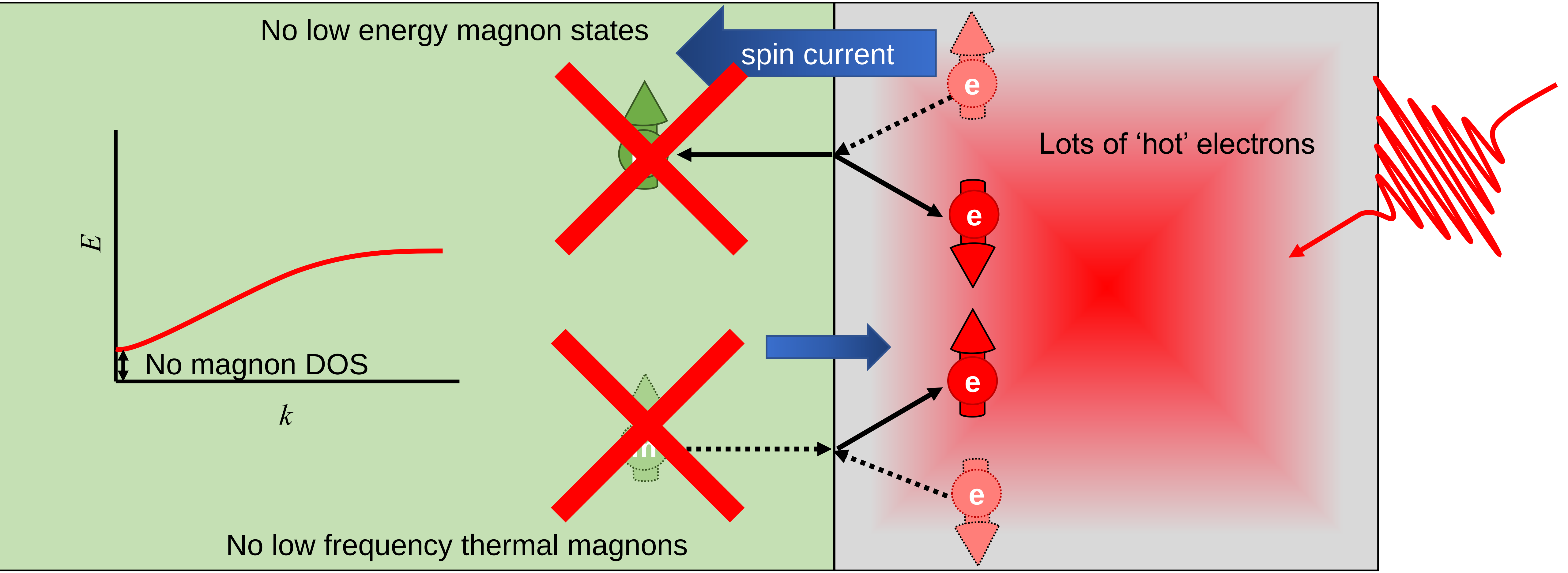
Interface spin currents – mDOS gap

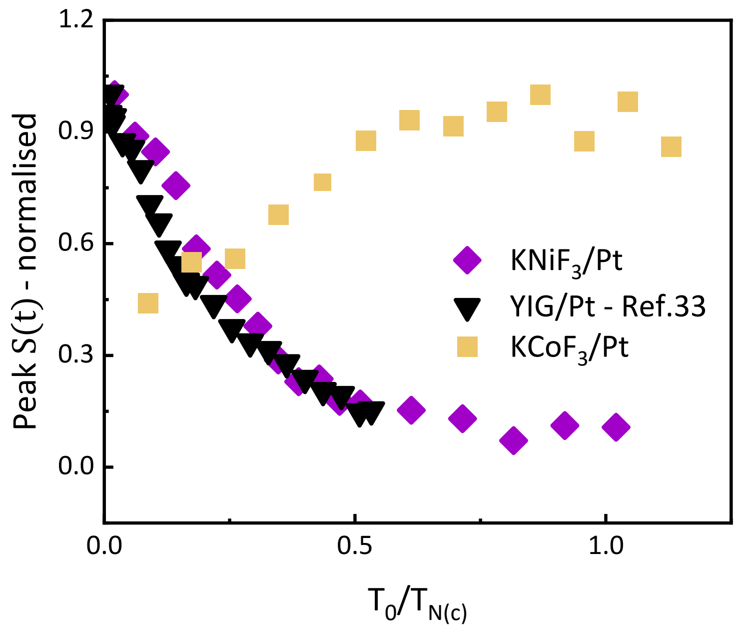
Interface spin current transfer is constrained by the lack of magnon states.
Laser only increases Pt electron temperature ~30K, so at low temperatures few electrons can spin transfer



Still some open questions

- No theory yet which includes both spin transfer torques and incoherent processes – maybe important to understand the role of different domains in antiferromagnets.

- Small gap ferromagnets and antiferromagnets seem to have a universal behaviour. Not clear why.



Conclusions

- THz spin currents can be generated by antiferromagnet/heavy metal bilayers and they probe the interface spin-charge conversion.

- Experimental results are well reproduced by a simple sd-scattering model.

- Spin current behaviour gives information about gaps in the magnon density of states.

Thank You