Towards coupling coherent femtosecond charge and spin dynamics

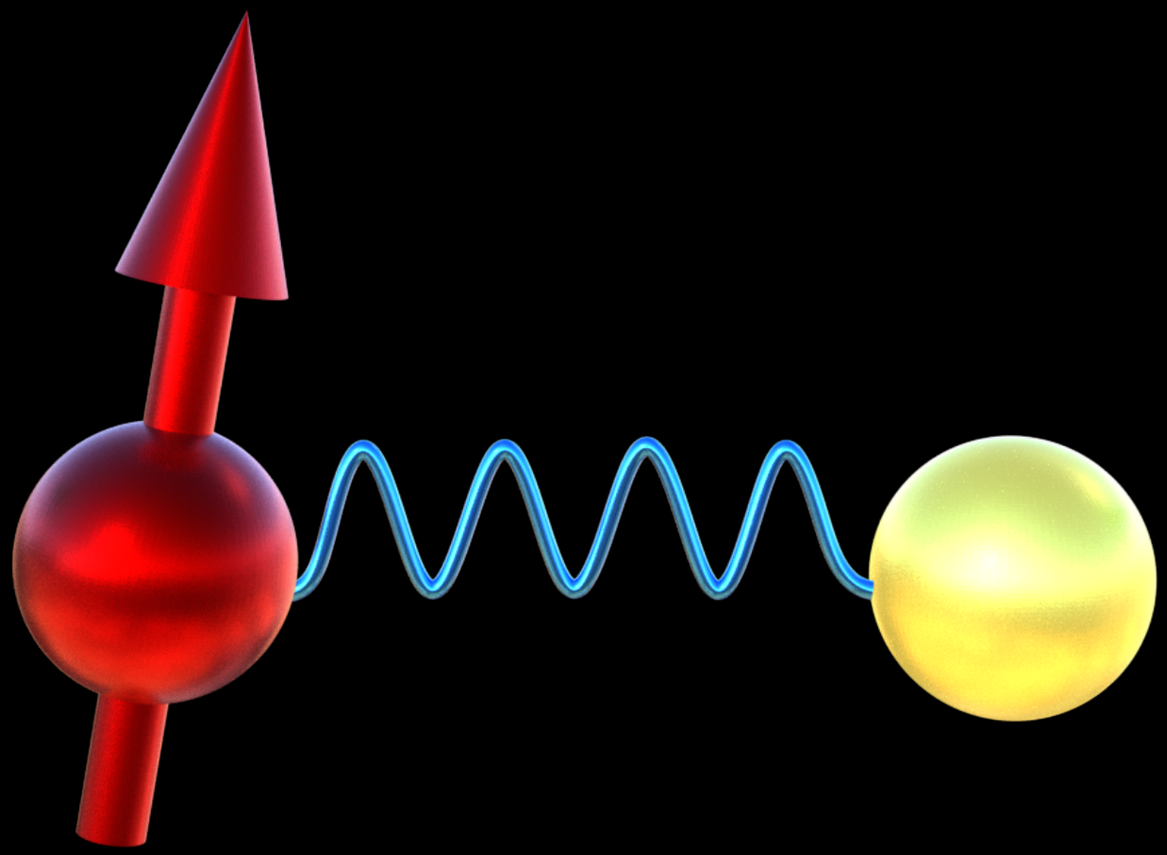
D. Bossini

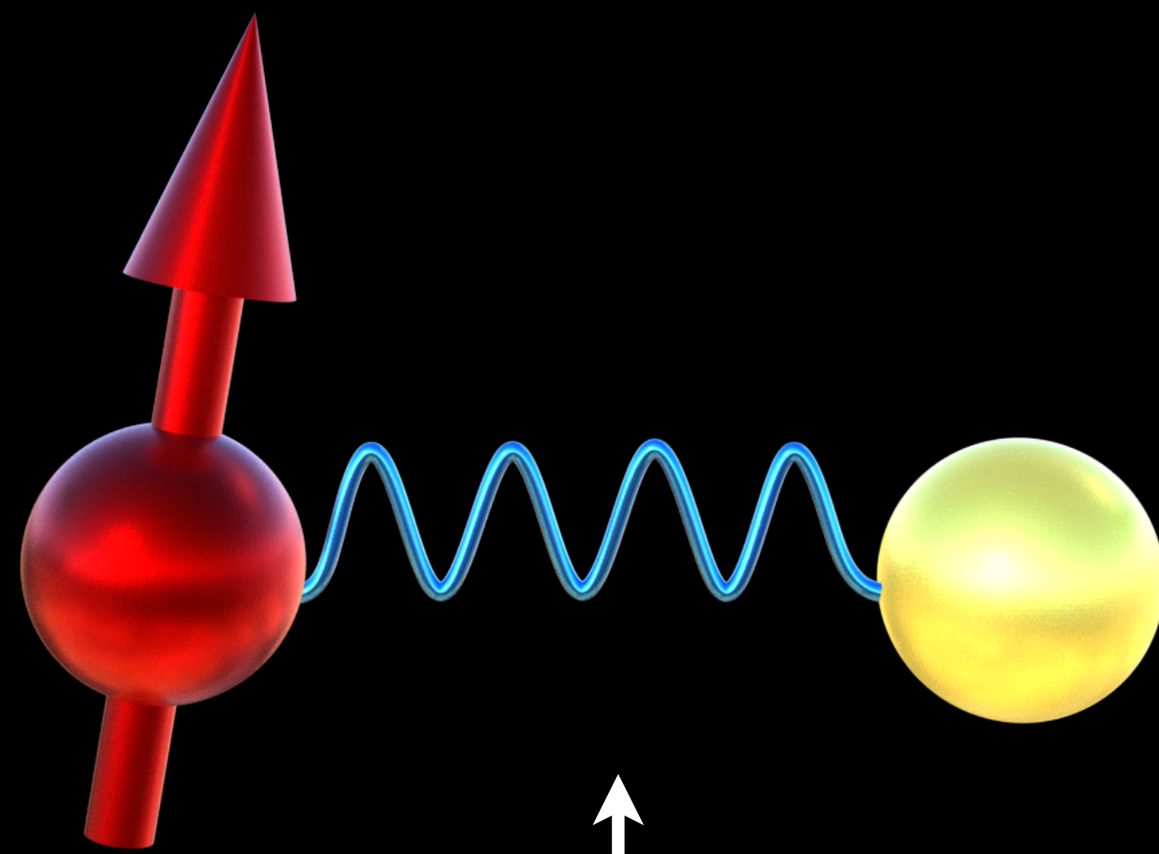
Department of Physics, University of Konstanz, Germany

Emmy Noether-Programm

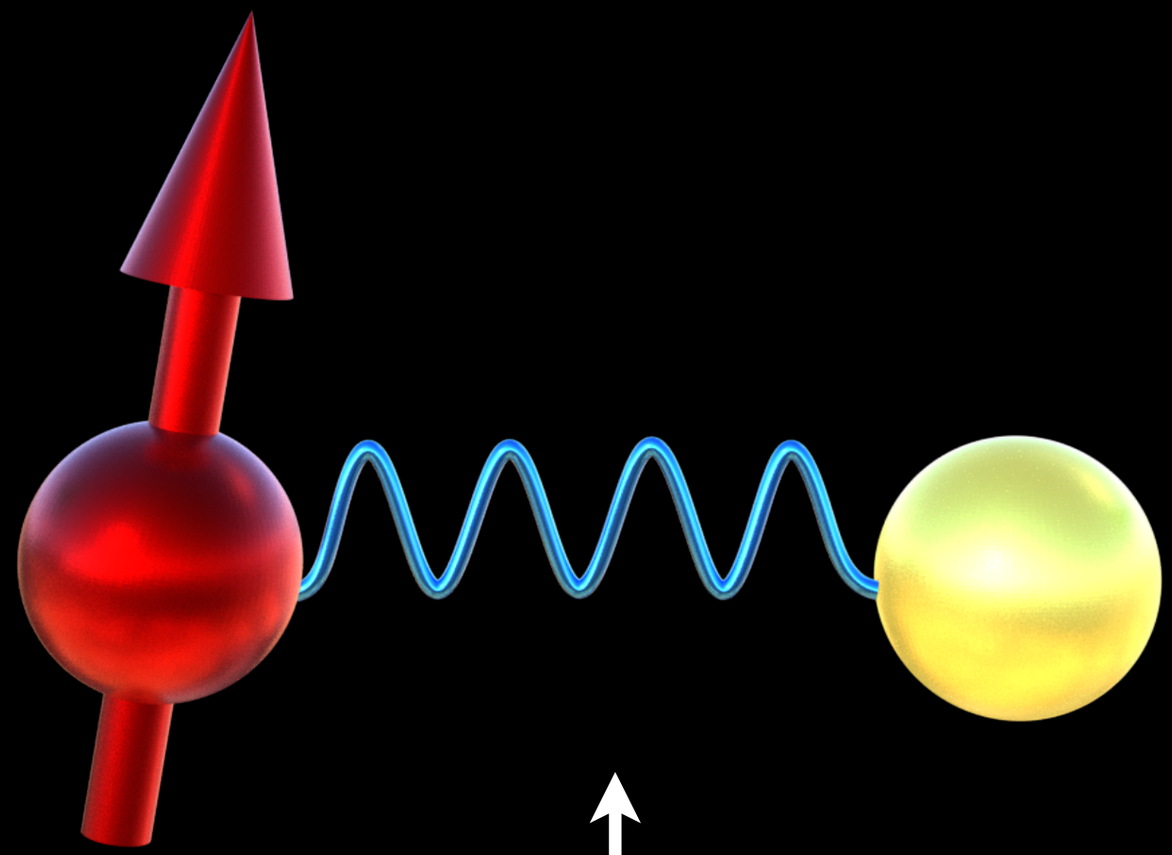
DFG Deutsche Forschungsgemeinschaft

Universität Konstanz






1 ns
1 ps
1 fs



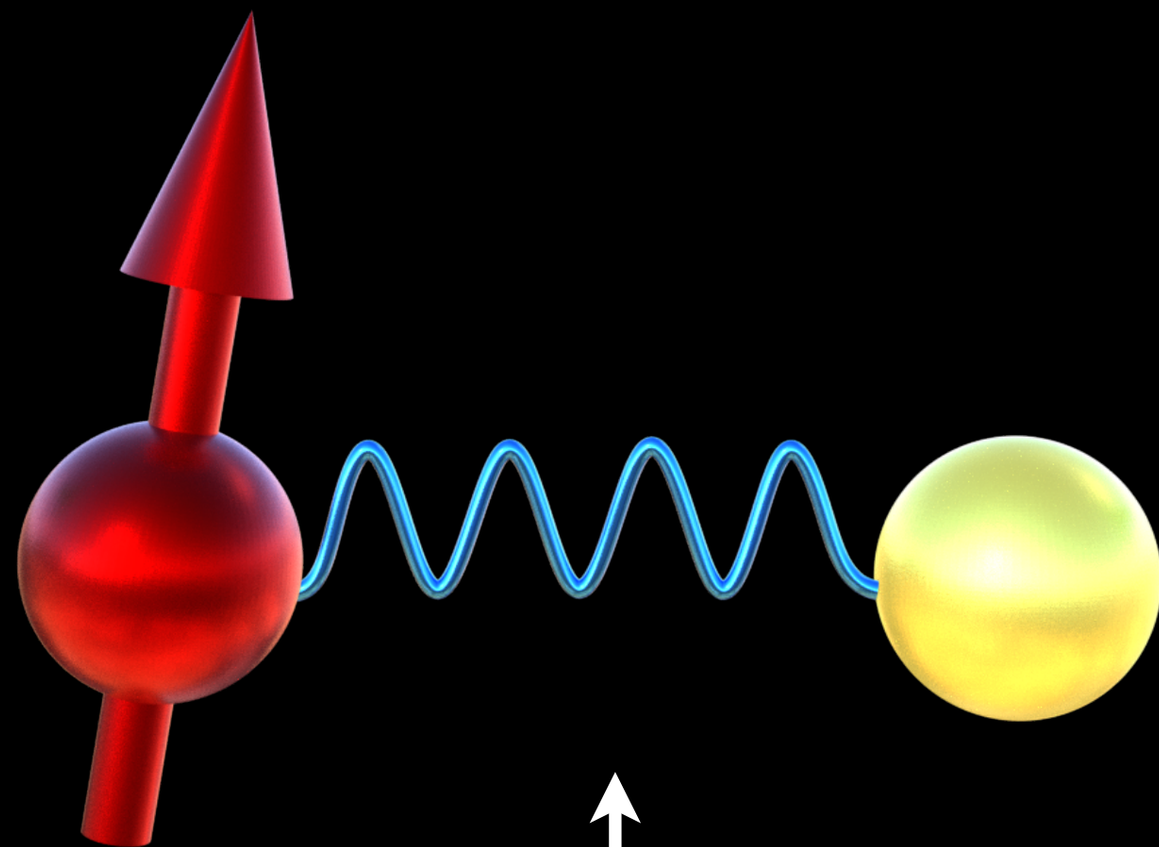
Science

adiabatic approx
equilibrium

1 ns

1 ps

1 fs



Science

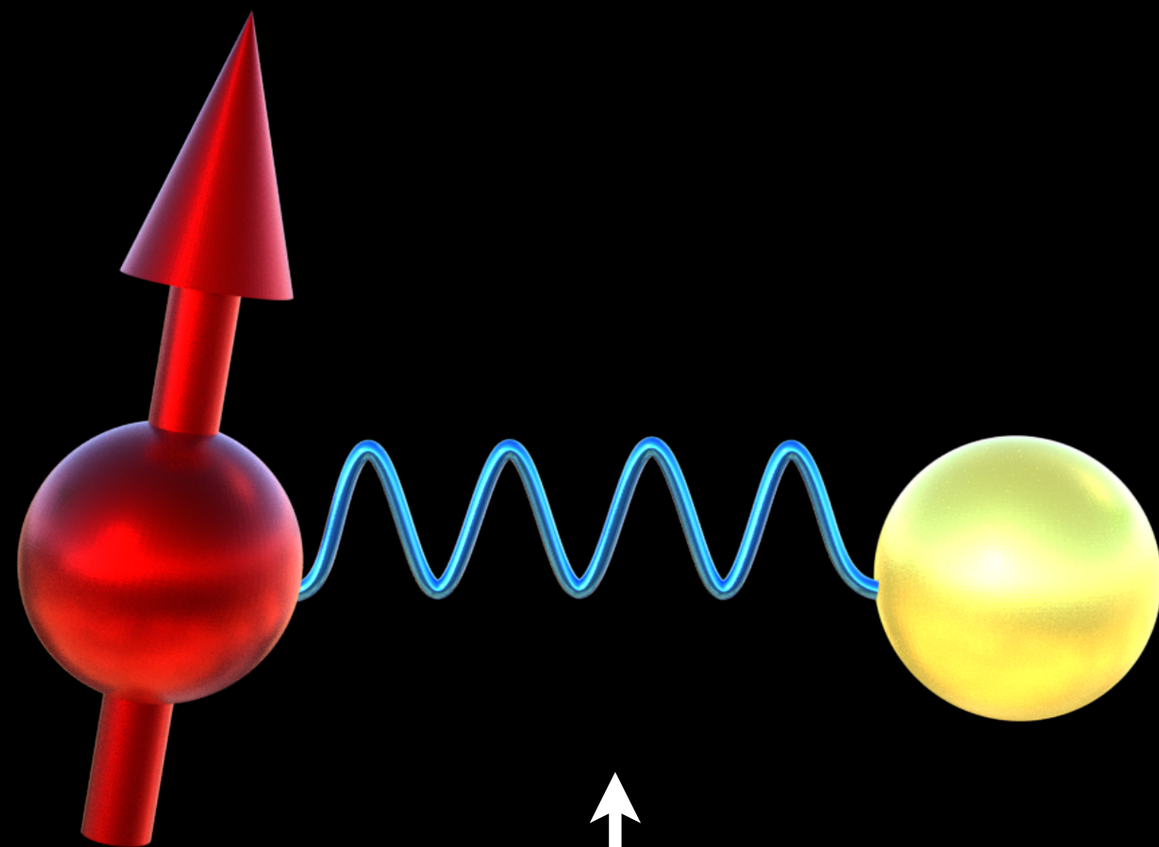
adiabatic approx

~~equilibrium~~

1 ns

1 ps

1 fs



Science

~~adiabatic approx~~

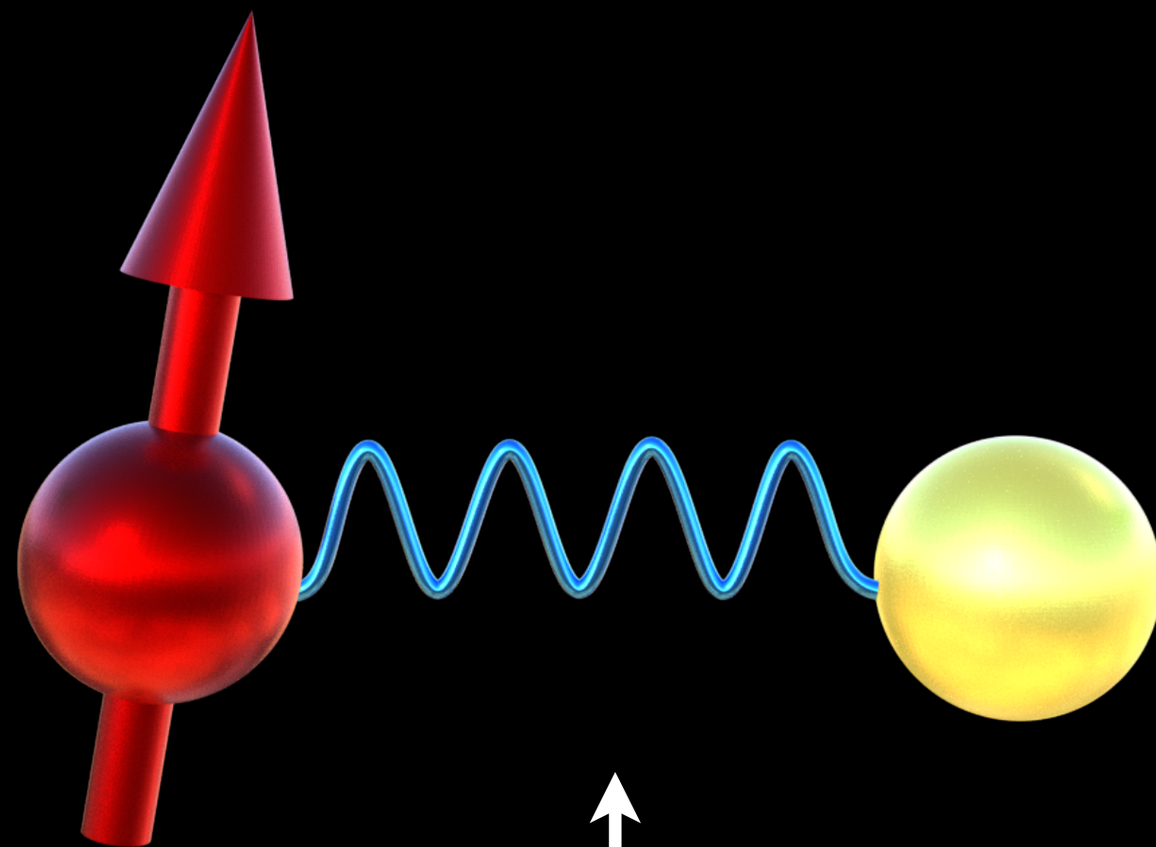
~~equilibrium~~

classical ⟷ quantum

1 ns

1 ps

1 fs



Science

adiabatic approx

equilibrium

classical ⟷ quantum

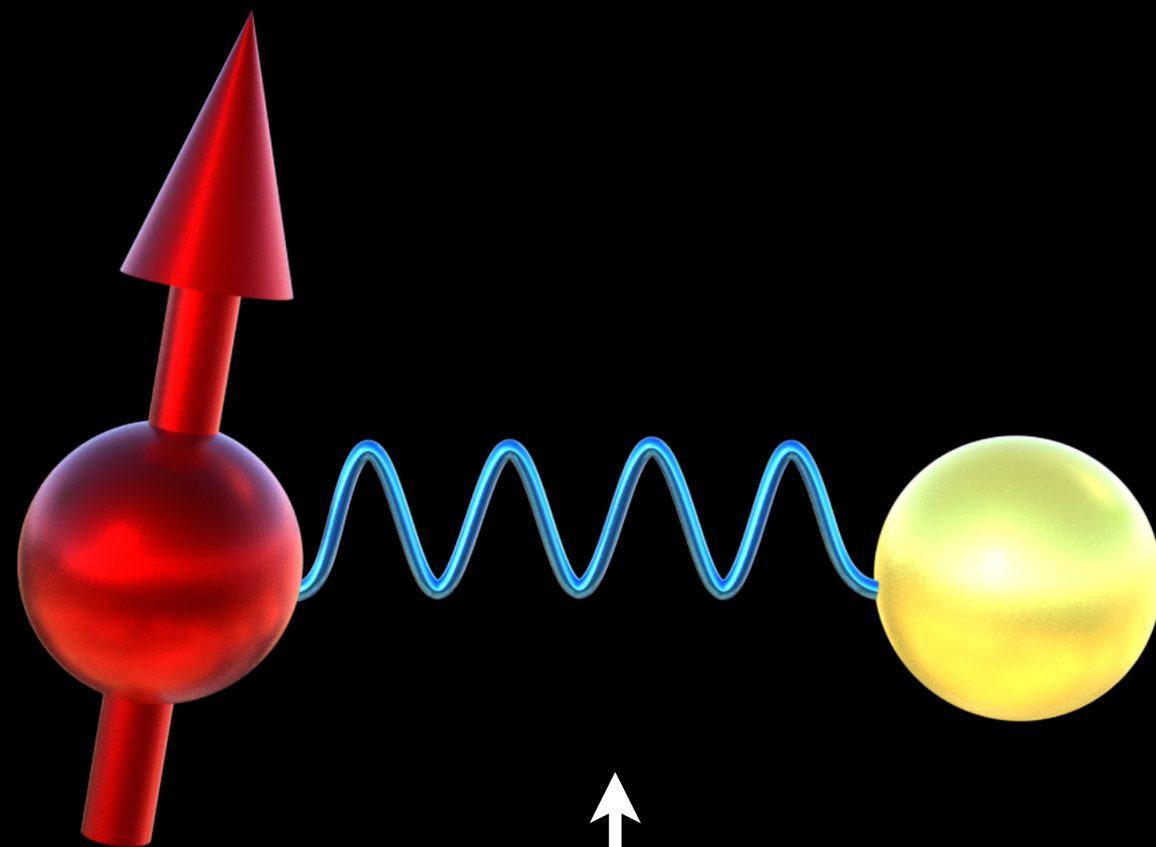
mean field

equations of motion

1 ns

1 ps

1 fs



Science

~~adiabatic approx~~

~~equilibrium~~

classical ⟷ quantum

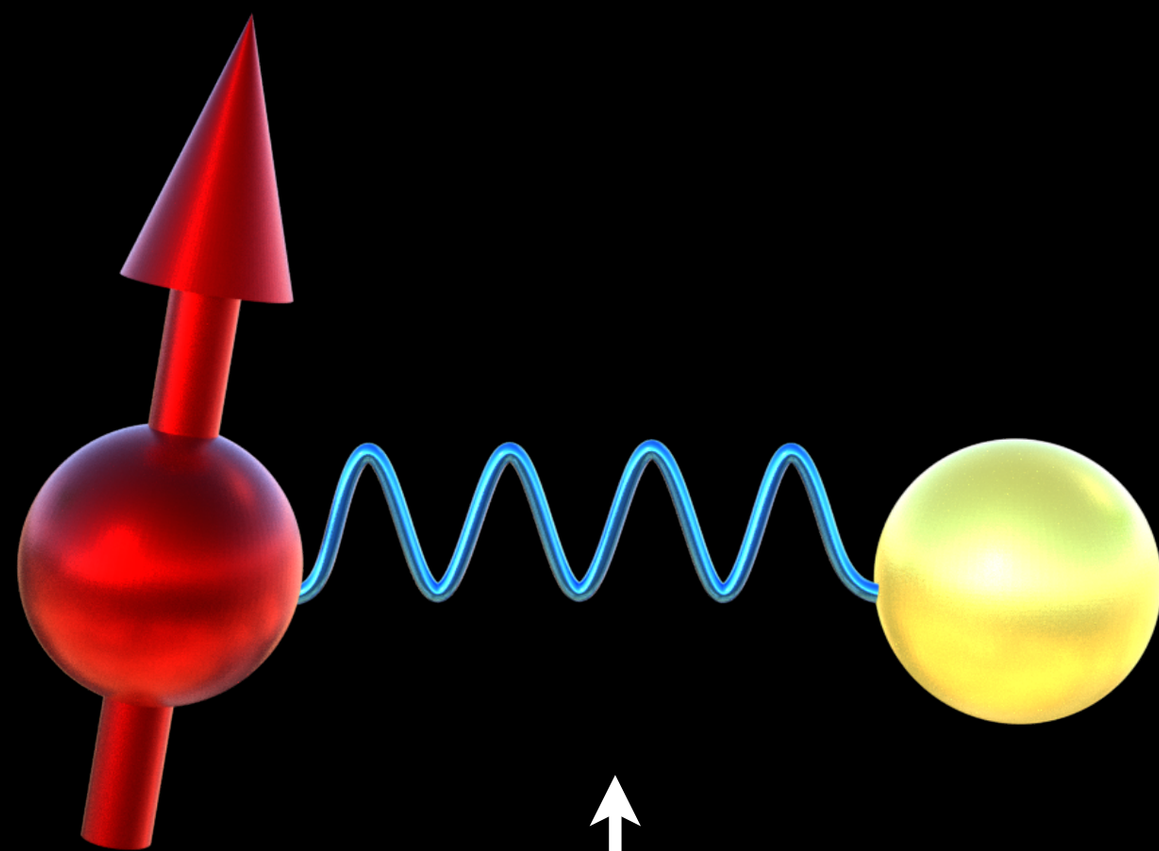
~~mean field~~

~~equations of motion~~

1 ns

1 ps

1 fs



Science

adiabatic approx

equilibrium

classical ⟷ quantum

mean field

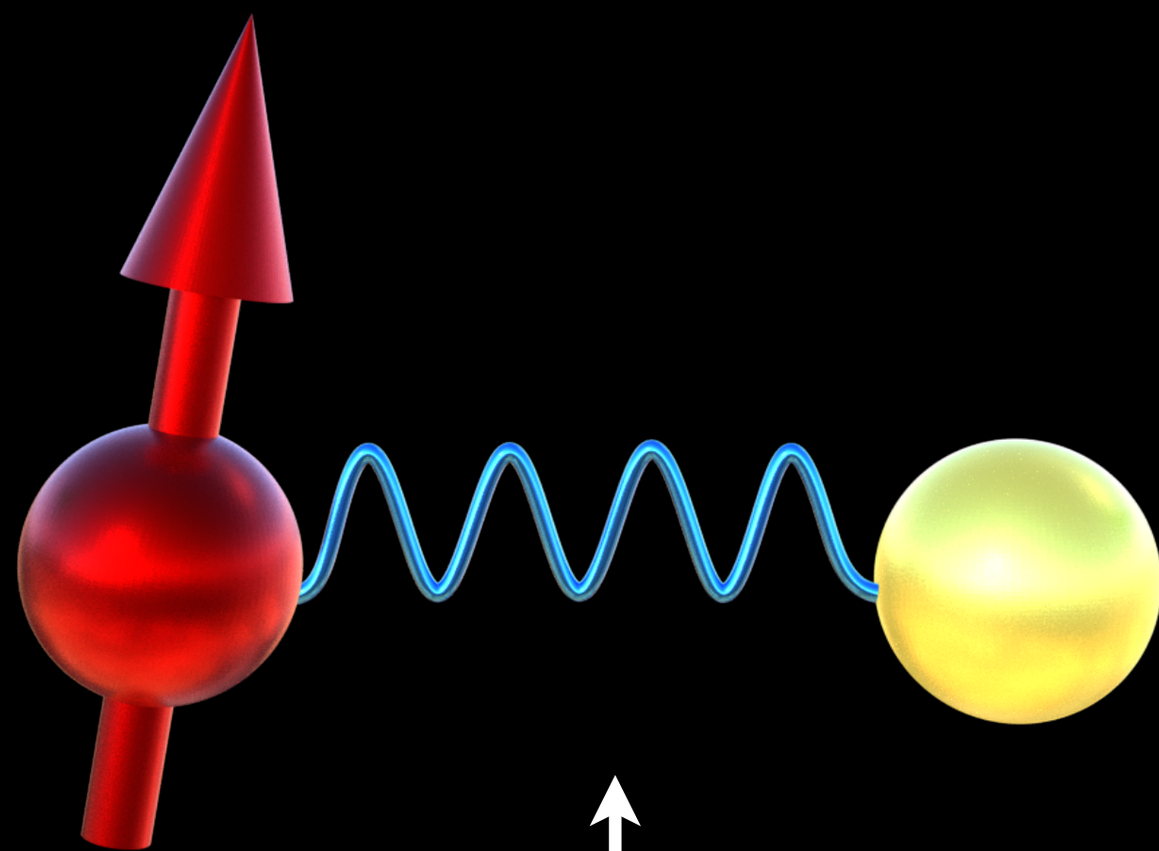
equations of motion

1 ns

1 ps

1 fs

Challenges



Science

1 ns

1 ps

1 fs

Challenges

~~adiabatic approx~~

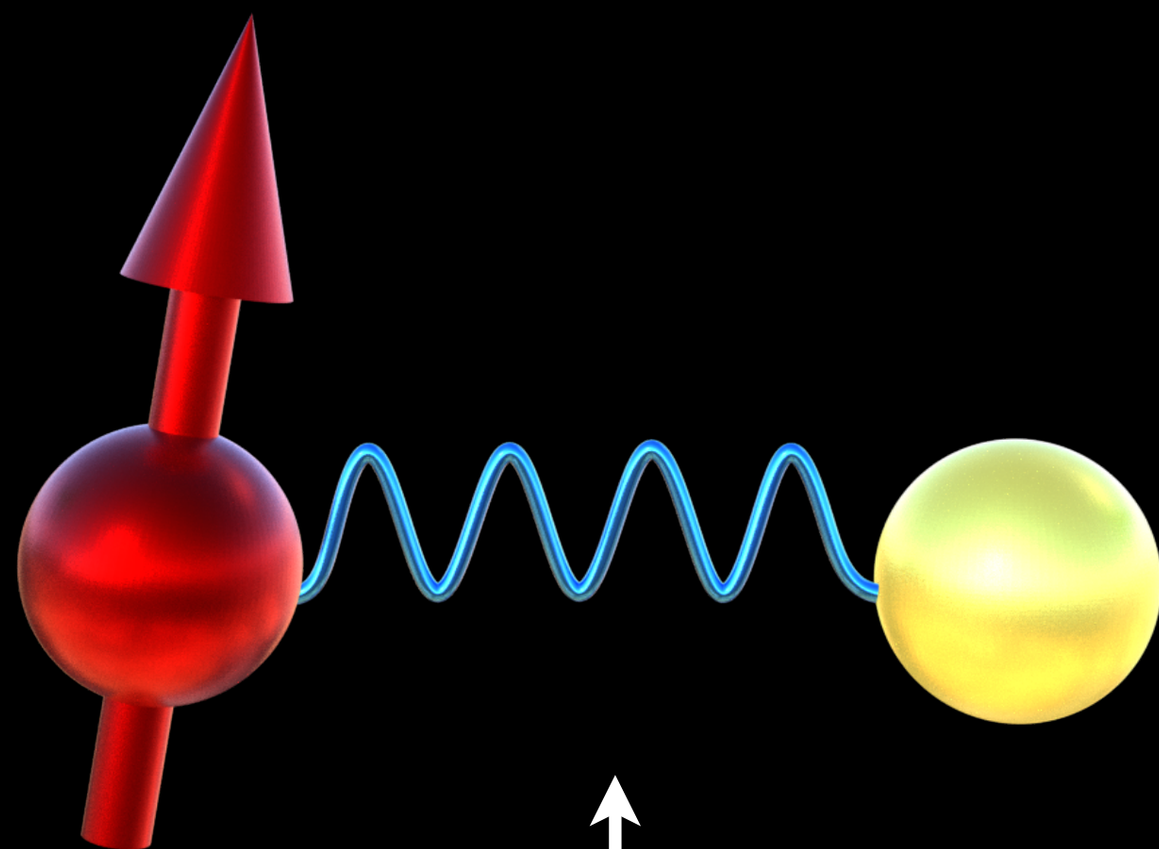
~~equilibrium~~

classical ⟷ quantum

~~mean field~~

~~equations of motion~~

Manipulation magnetism



Science

1 ns

1 ps

1 fs

Challenges

~~adiabatic approx~~

~~equilibrium~~

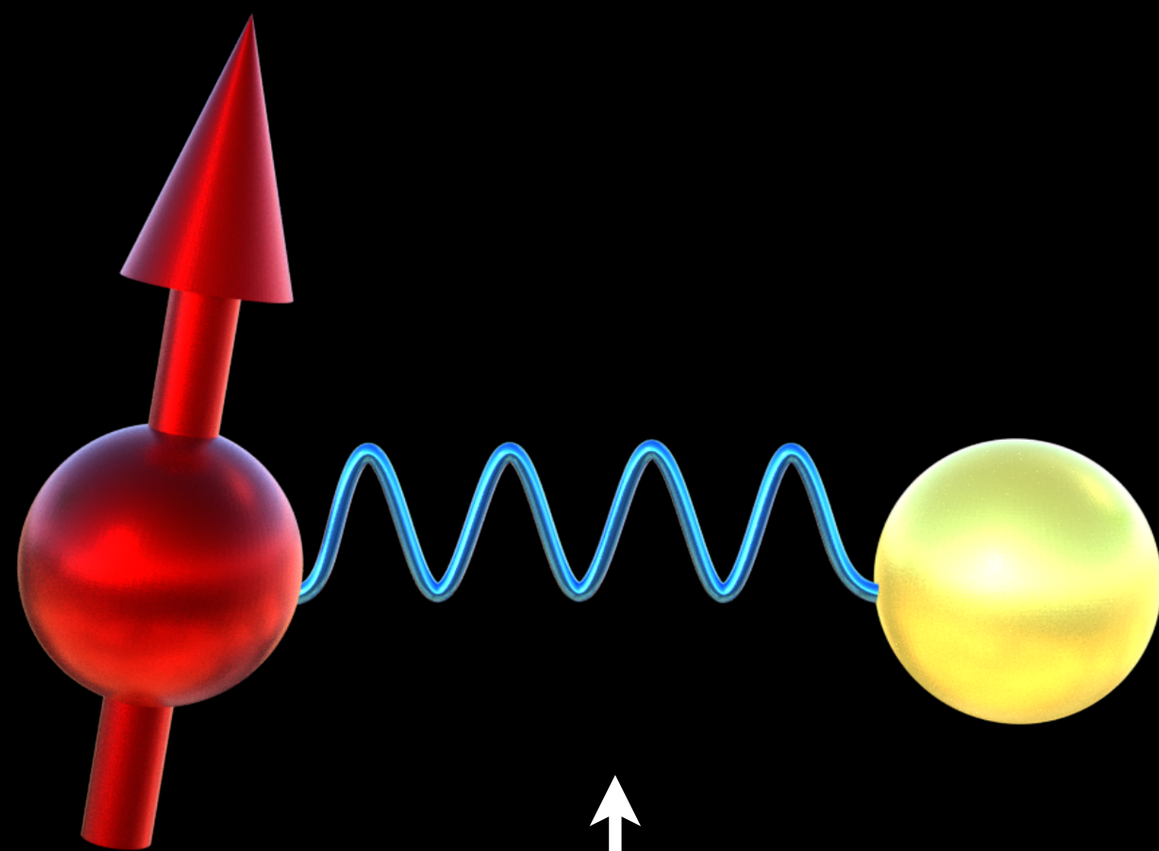
classical ⟷ quantum

~~mean field~~

~~equations of motion~~

Manipulation magnetism

Low dissipations: **coherence**



Science

1 ns

1 ps

1 fs

Challenges

~~adiabatic approx~~

~~equilibrium~~

classical ⟷ quantum

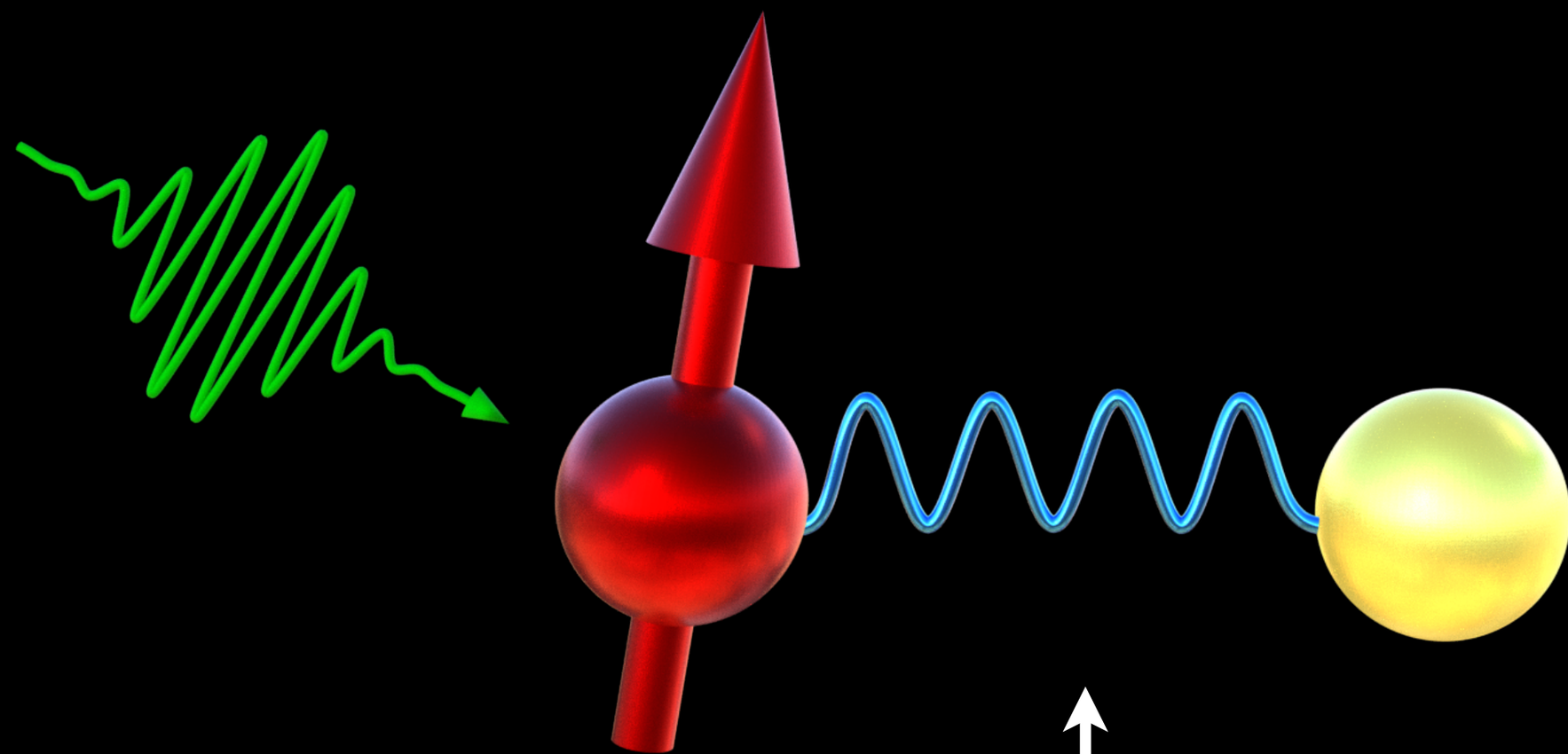
~~mean field~~

~~equations of motion~~

Manipulation magnetism

Low dissipations: **coherence**

Coupling to charges



Science

1 ns

1 ps

1 fs

Challenges

~~adiabatic approx~~

~~equilibrium~~

classical ⟷ quantum

~~mean field~~

~~equations of motion~~

Manipulation magnetism

Low dissipations: **coherence**

Coupling to charges

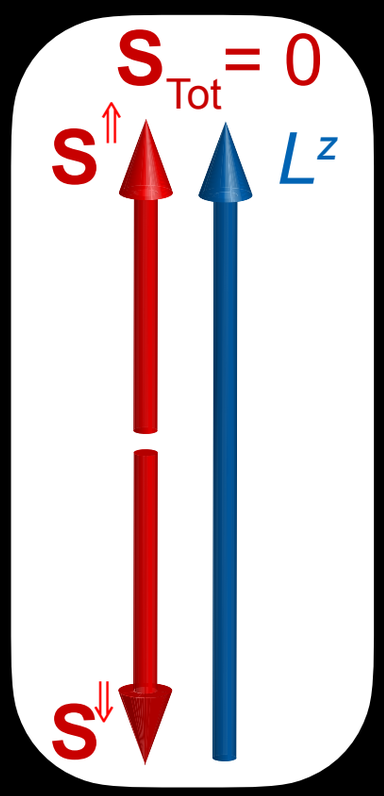
Dielectric antiferromagnet

- No free electrons
- Majority of ordered materials
- Intrinsically faster spin dynamics

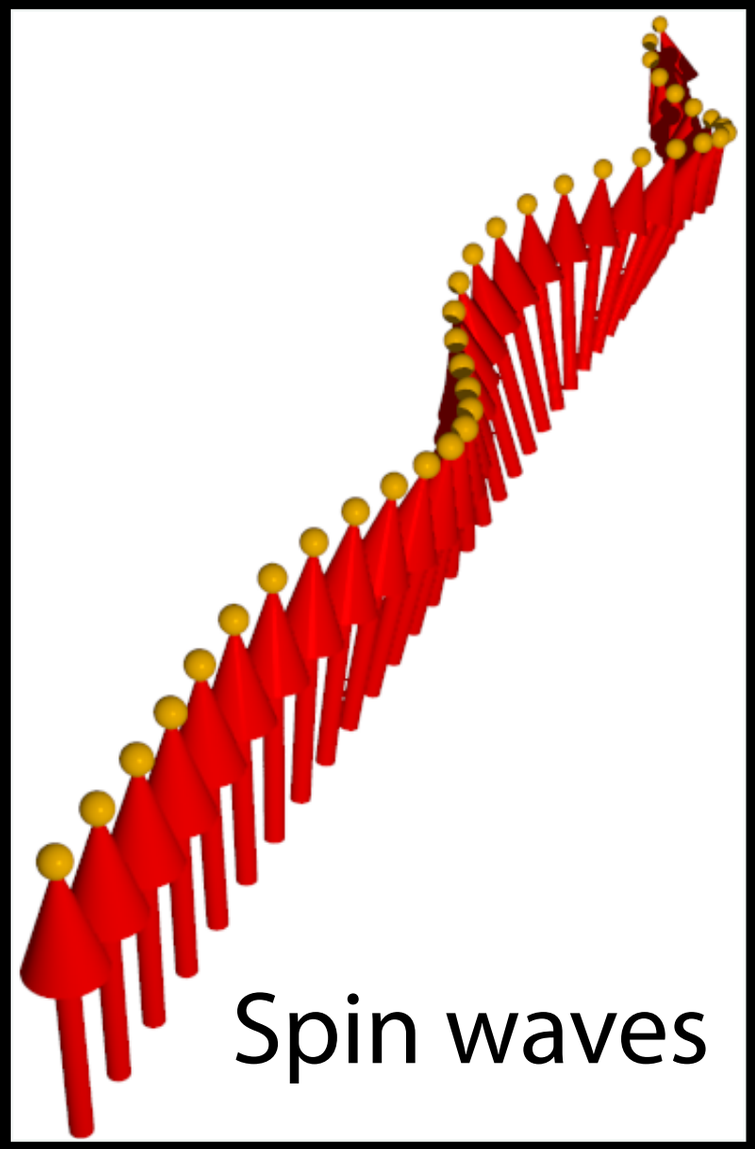
Dielectric antiferromagnet

- No free electrons
- Majority of ordered materials
- Intrinsically faster spin dynamics

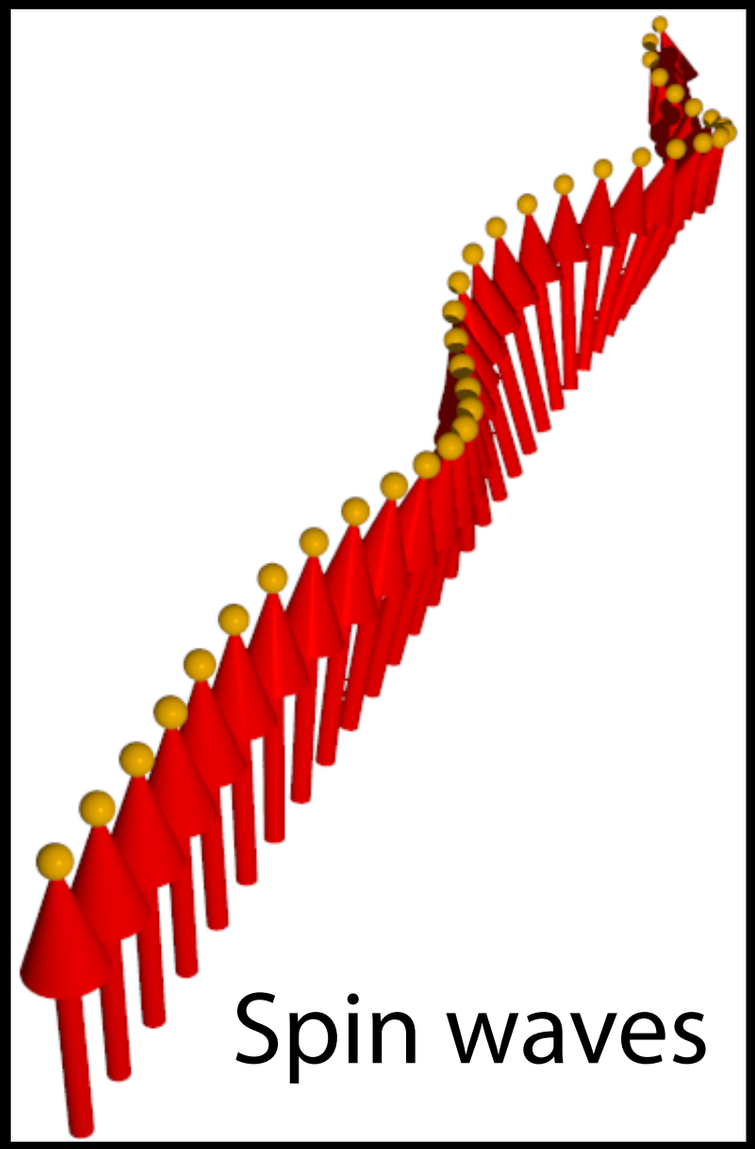
$$\boldsymbol{L} \equiv \boldsymbol{S}^{\Uparrow} - \boldsymbol{S}^{\Downarrow}$$



Magnon generation

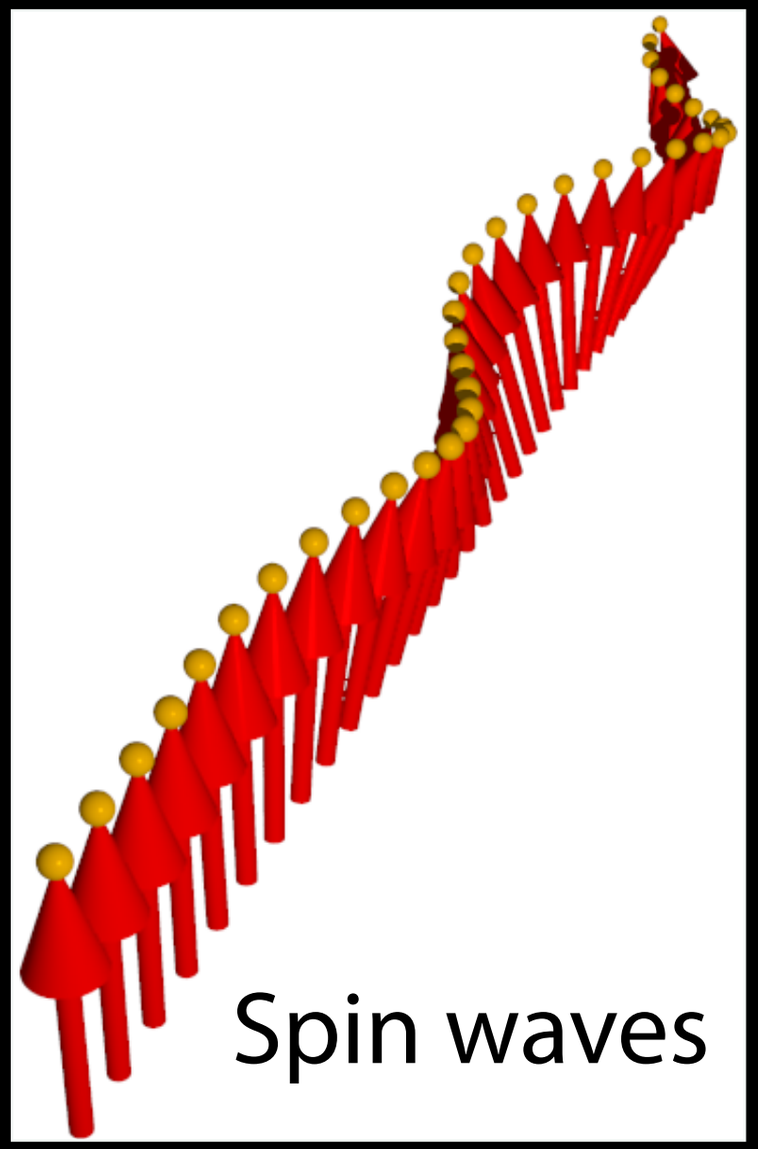


Magnon generation



Magnon generation:
Spin-flip process, $\Delta S = 1$

Magnon generation

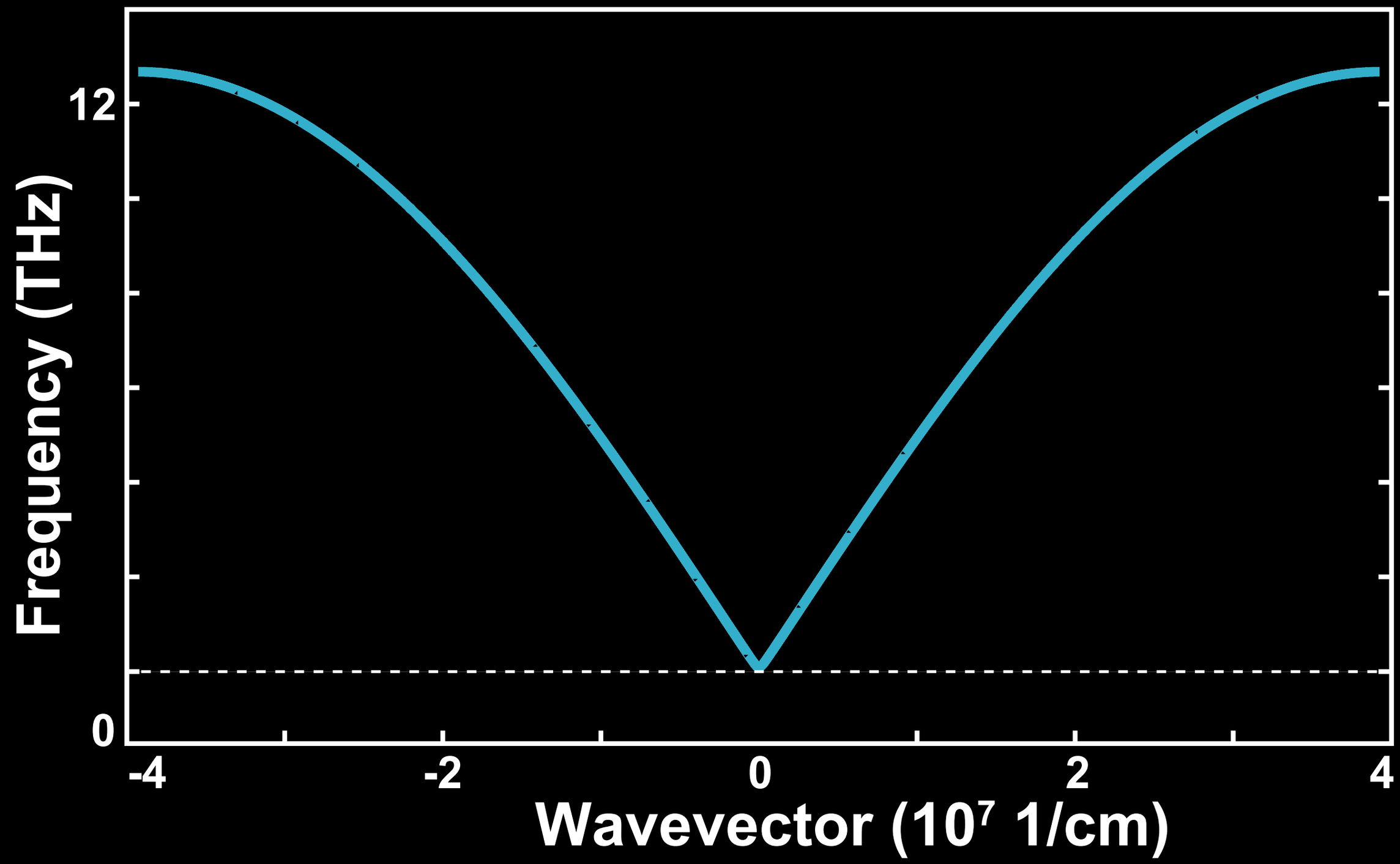


Magnon generation:
Spin-flip process, $\Delta S = 1$

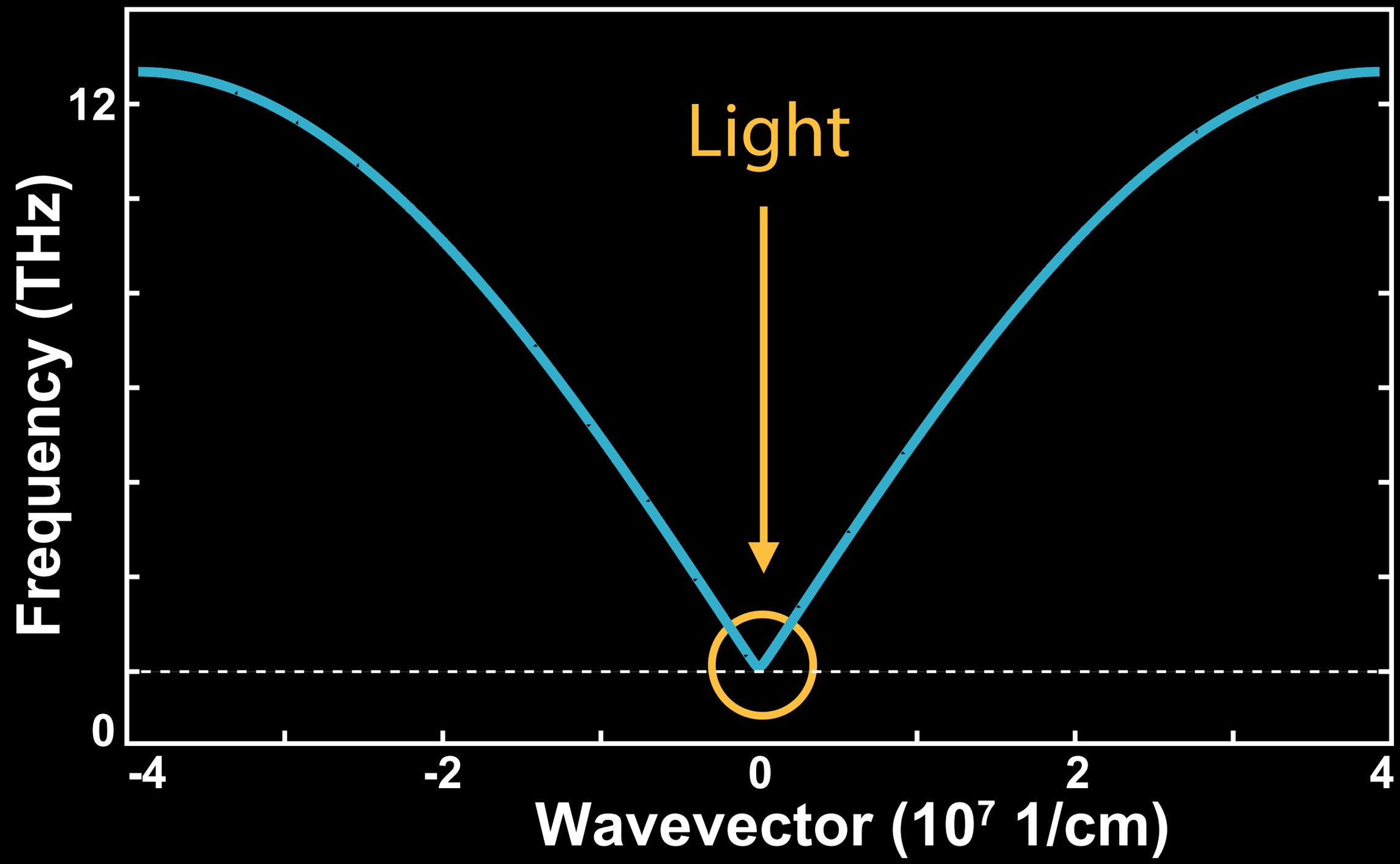
Spin-flip:

- Magnetic field
- Spin-orbit coupling

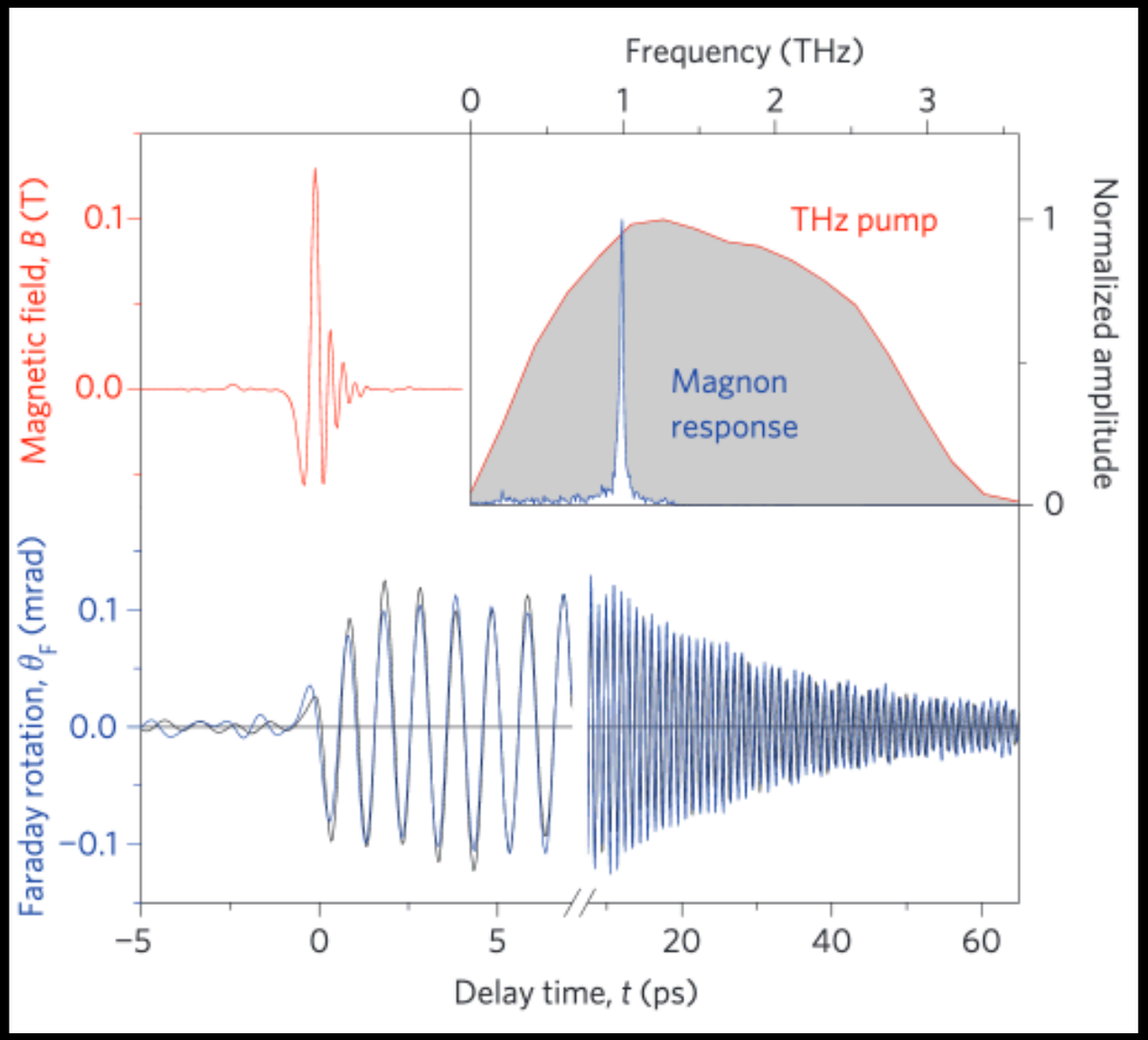
Magnon dispersion



Magnon dispersion

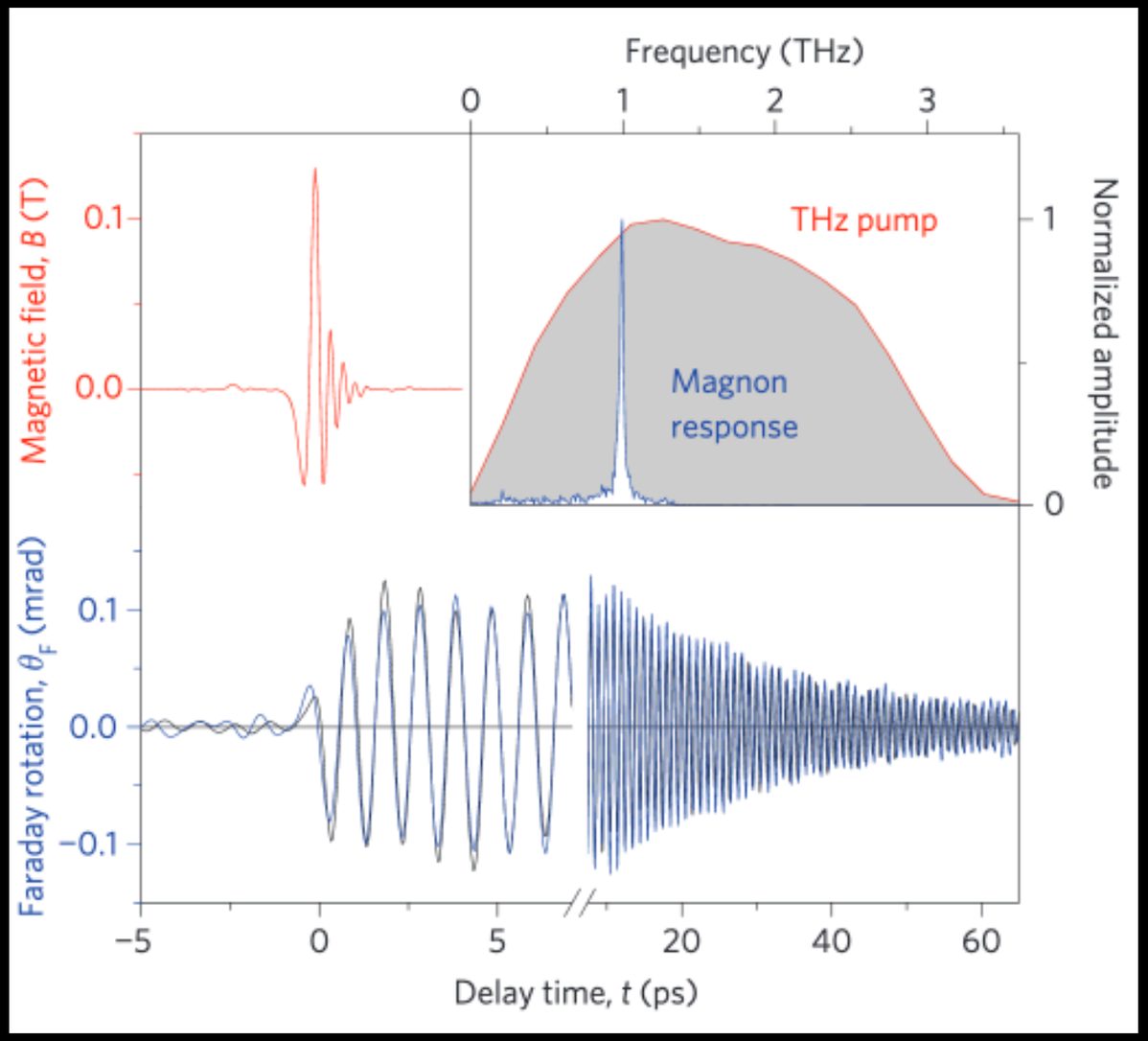


Resonant pump

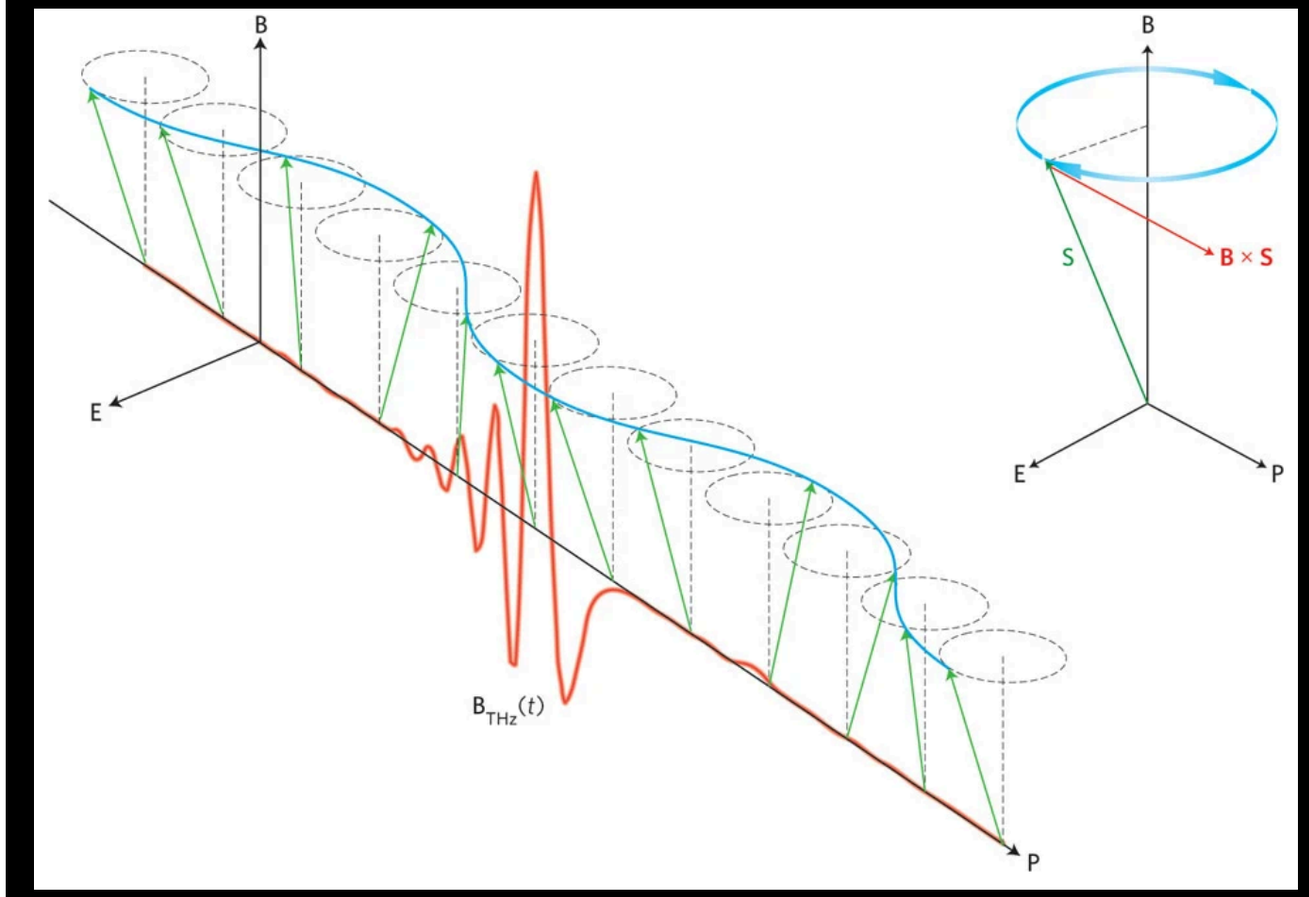


T. Kampfrath et al. Nat. Phot **5**, 31 (2011)

Resonant pump

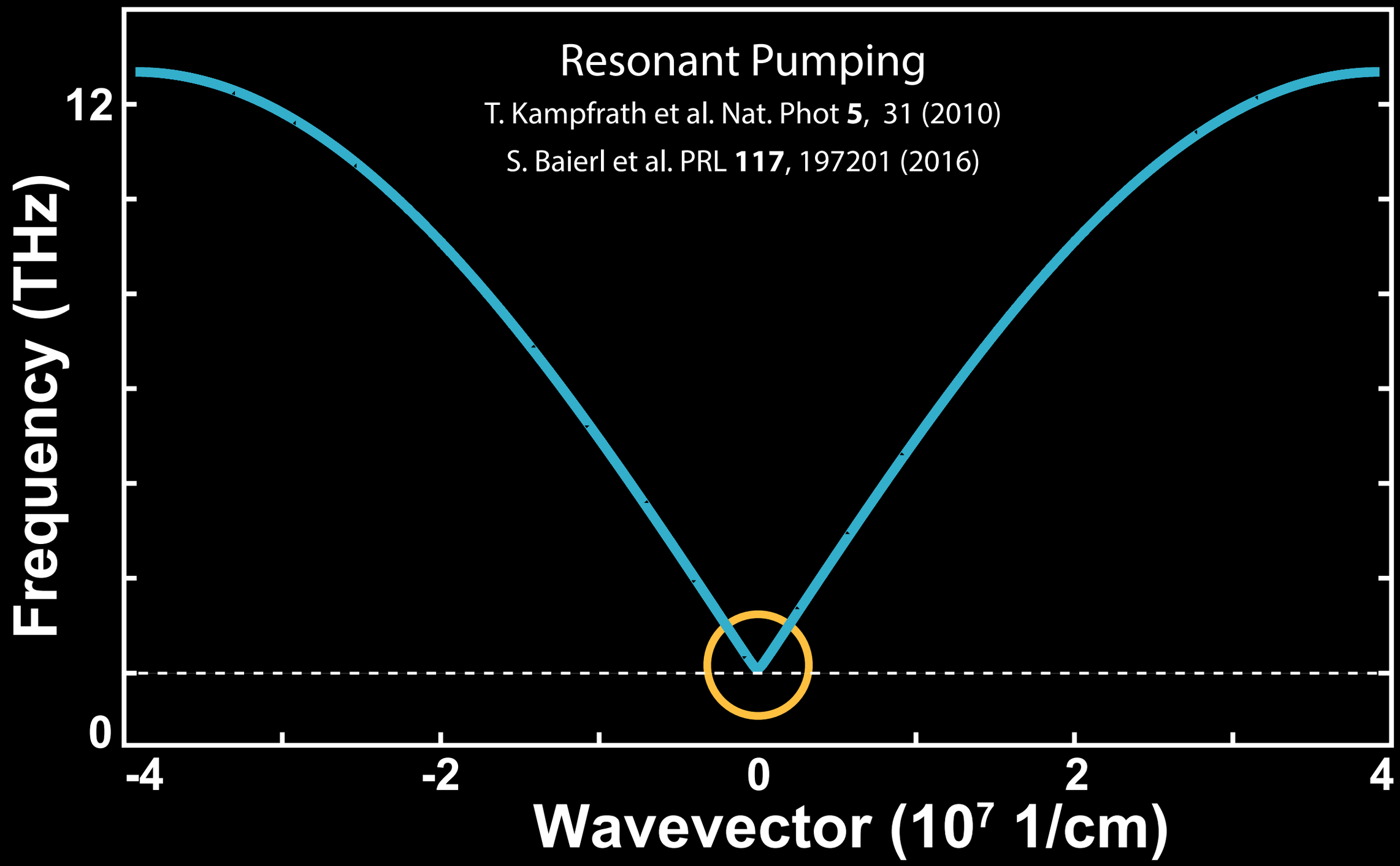


T. Kampfrath et al. Nat. Phot **5**, 31 (2011)

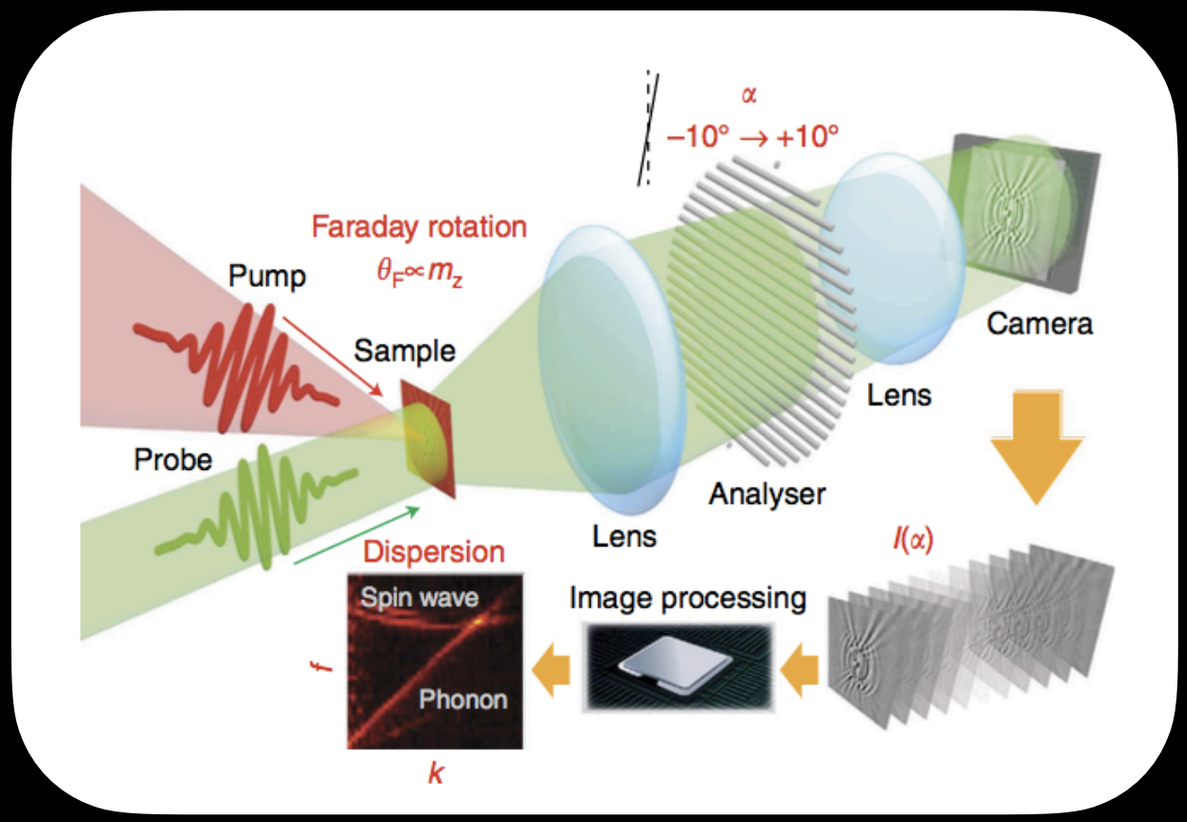


J. Kono Nat. Phot **5**, 5 (2011)

Resonant pump

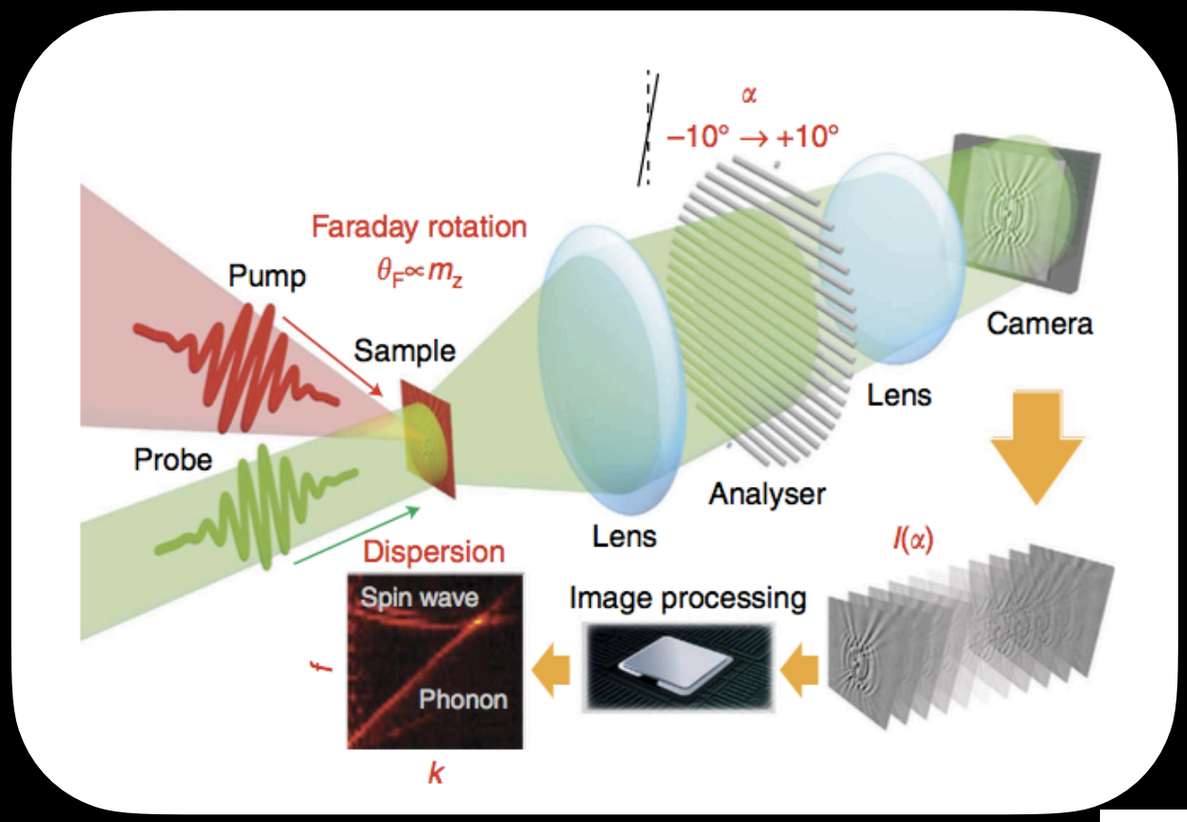


Magnetoelastic coupling



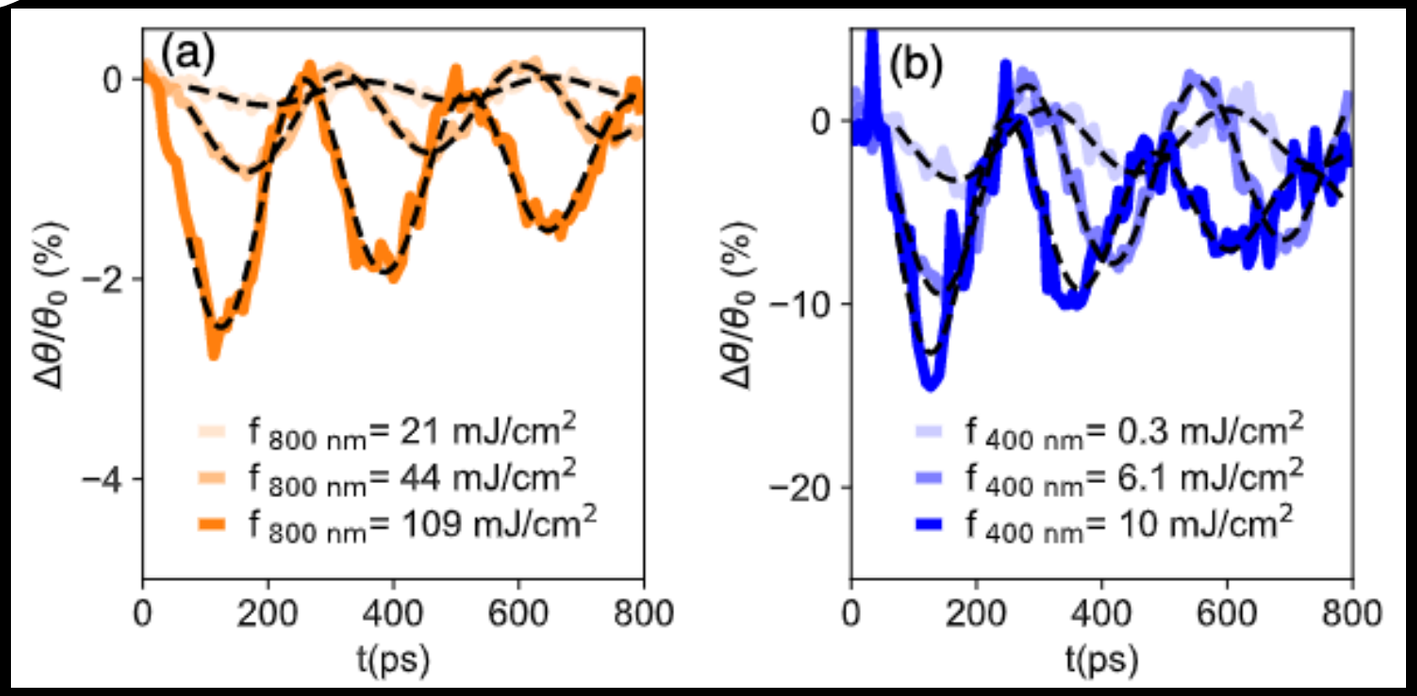
Y. Hashimoto, **DB** *et al.* Nat. Comm. **8,** 15859 (2017)

Magnetoelastic coupling

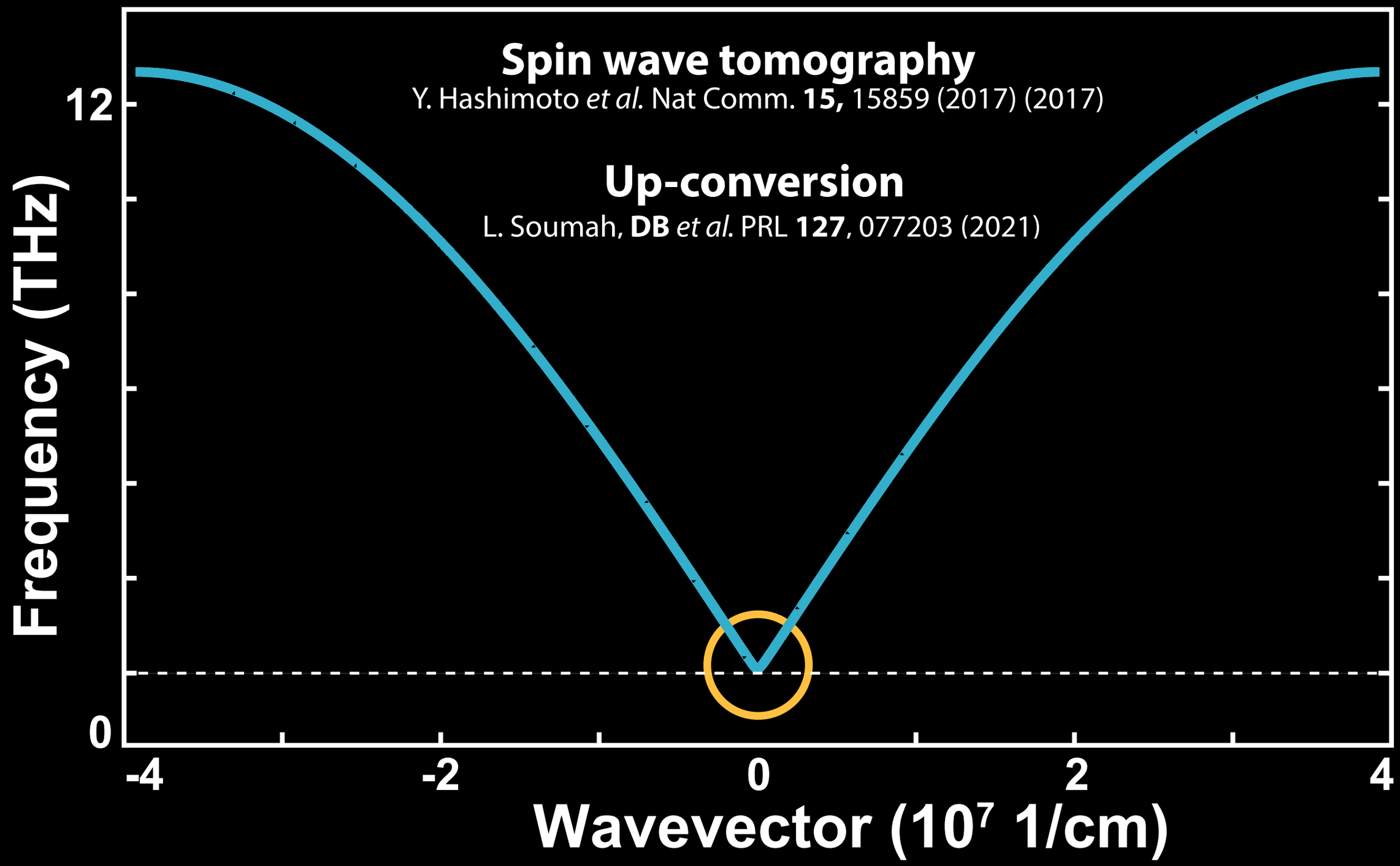


Y. Hashimoto, **DB** *et al.* Nat. Comm. **8,** 15859 (2017)

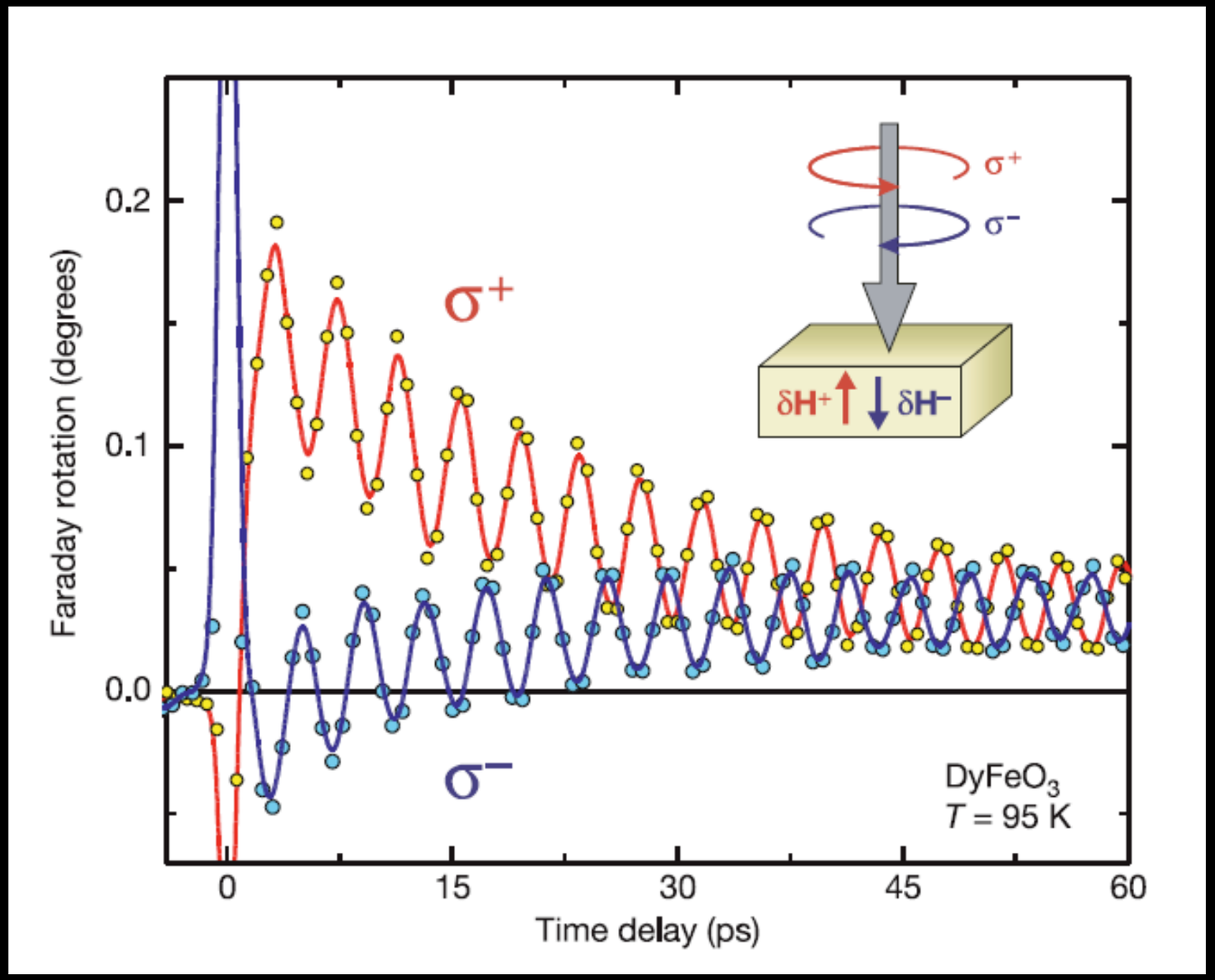
L. Soumah, **DB** *et al.* PRL **127**, 077203 (2021)



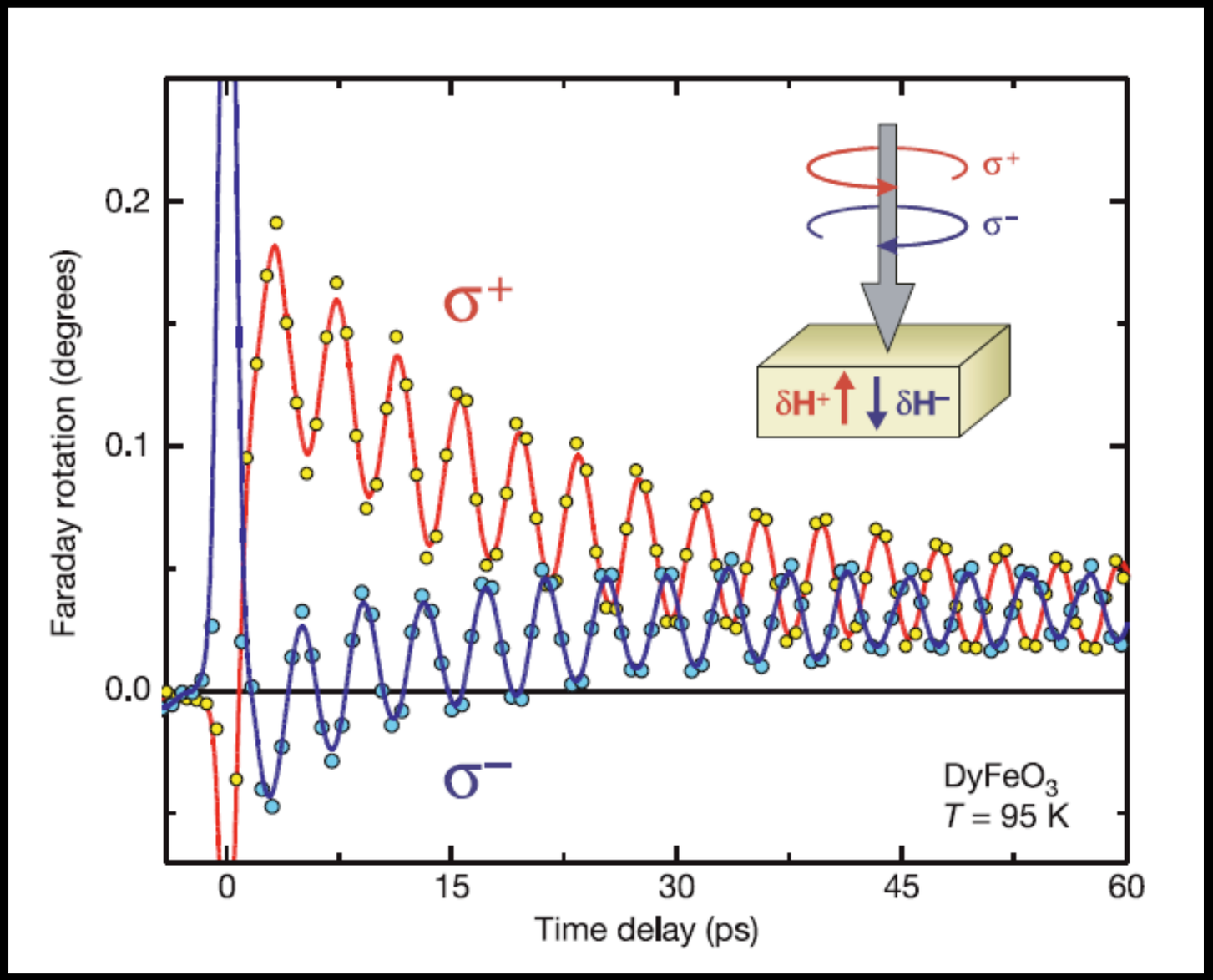
Magnon dispersion



A. Kimel *et al.* Nature **435,** 655 (2005)

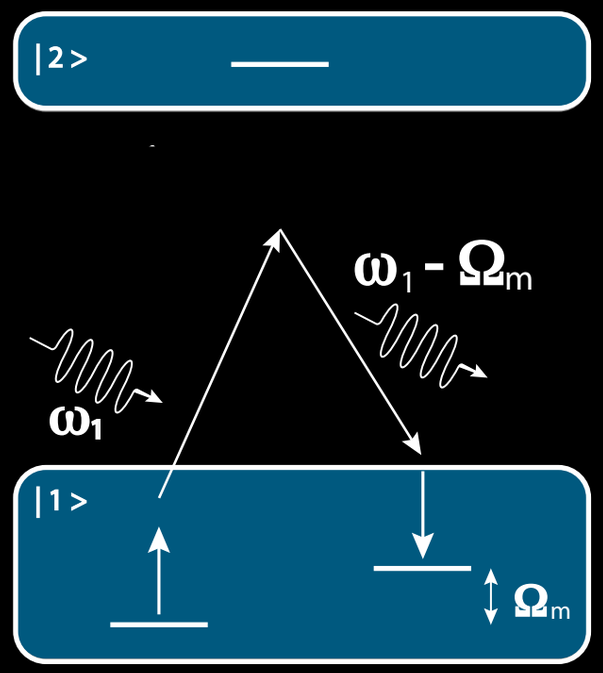


A. Kimel *et al.* Nature **435,** 655 (2005)

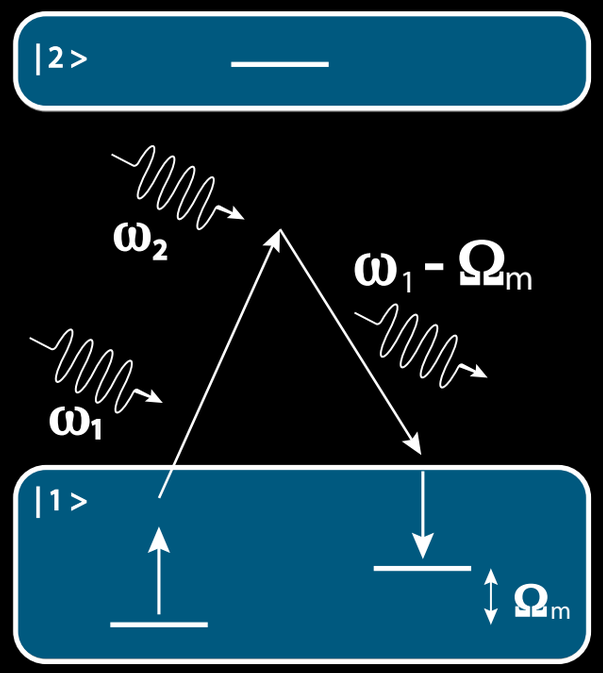


Impulsive stimulated Raman scattering (ISRS)

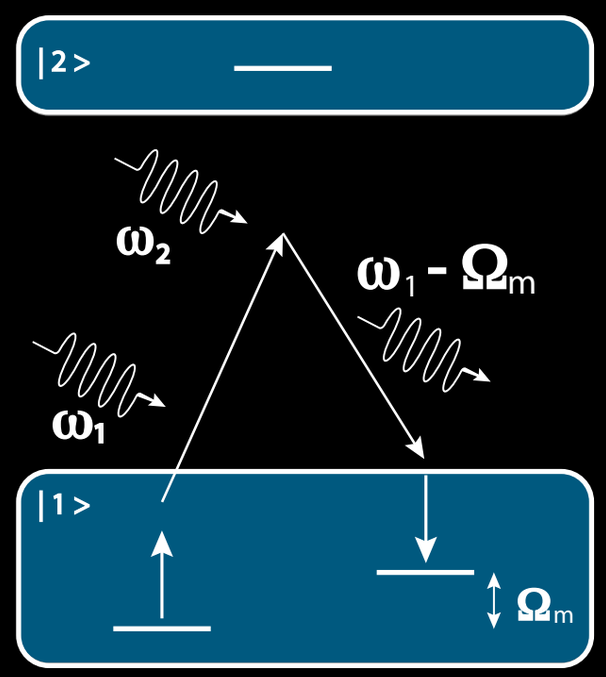
Light-scattering

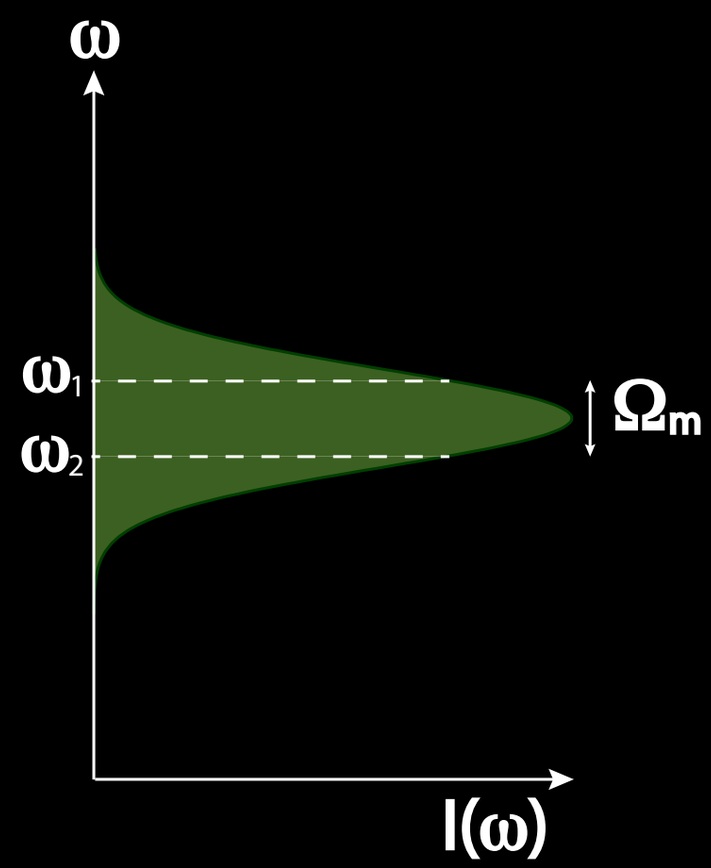


Light-scattering

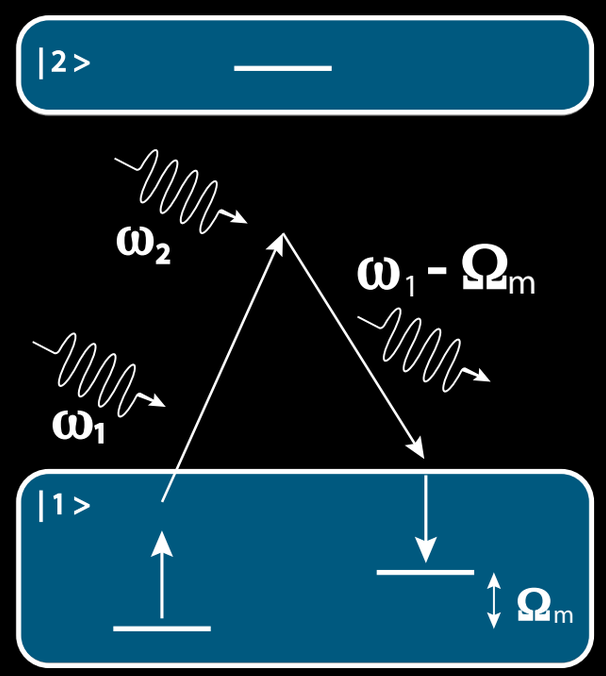


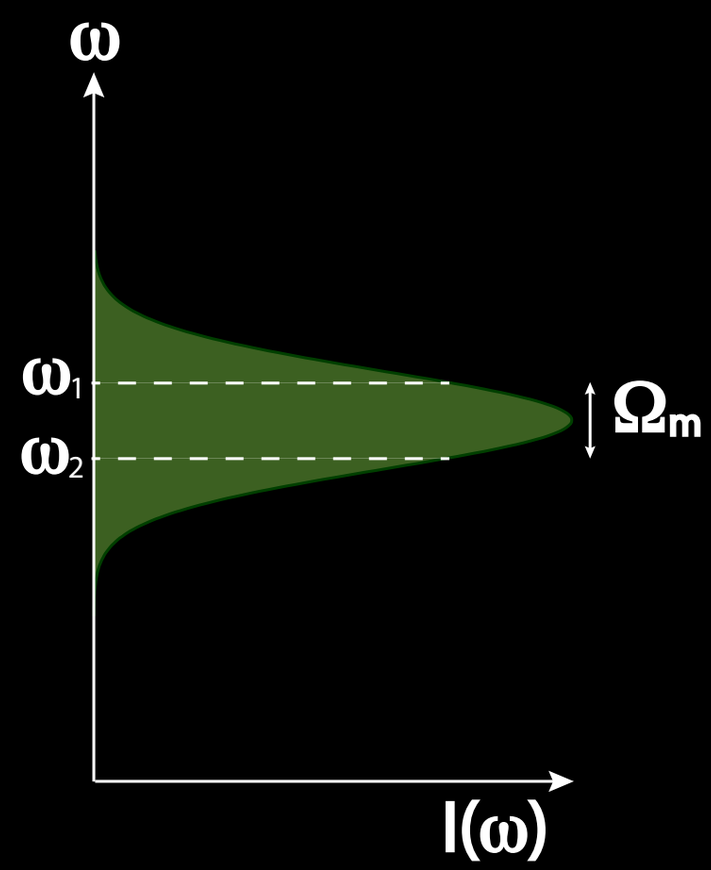
Light-scattering



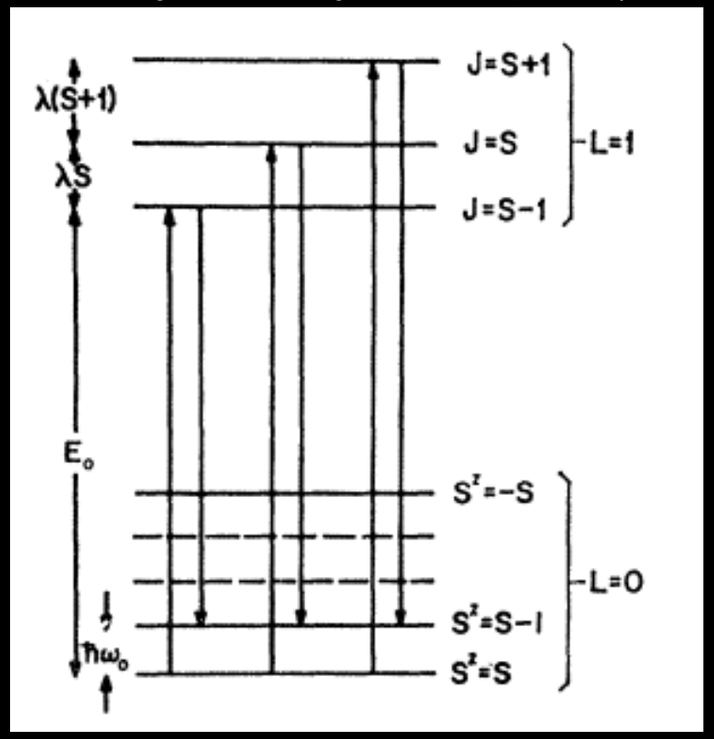


Light-scattering

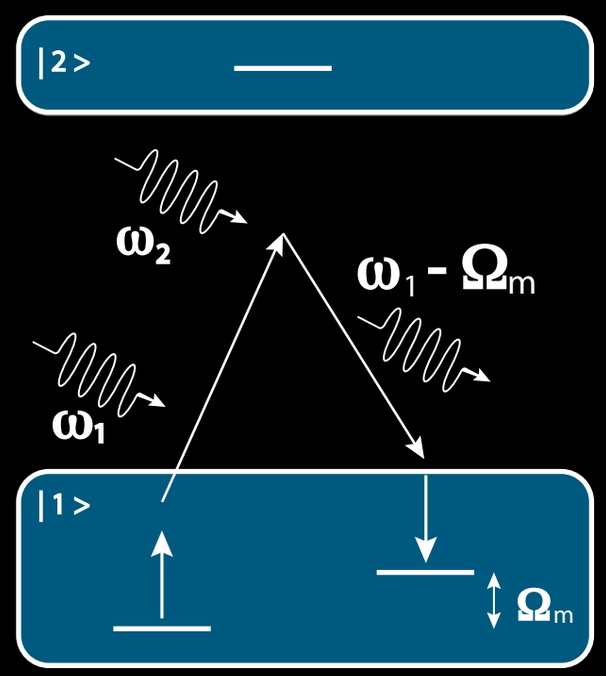


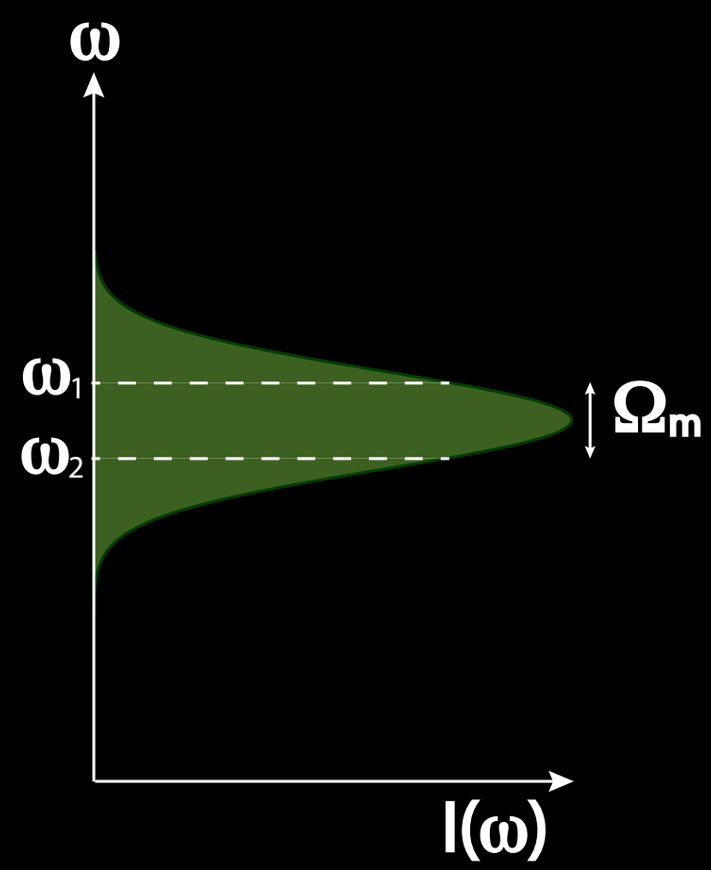


P. Fleury *et al.* Phys. Rev. **2,** 514 (1968)

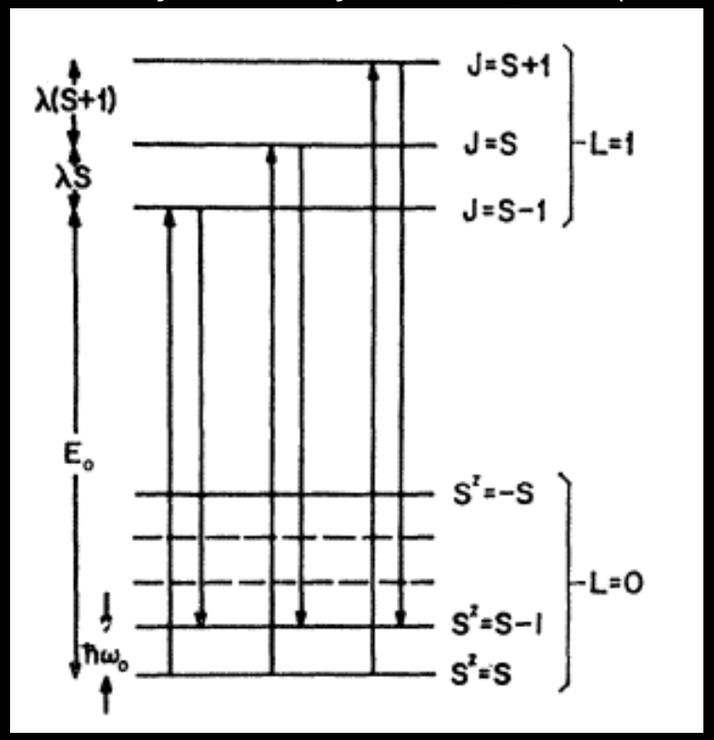


Light-scattering



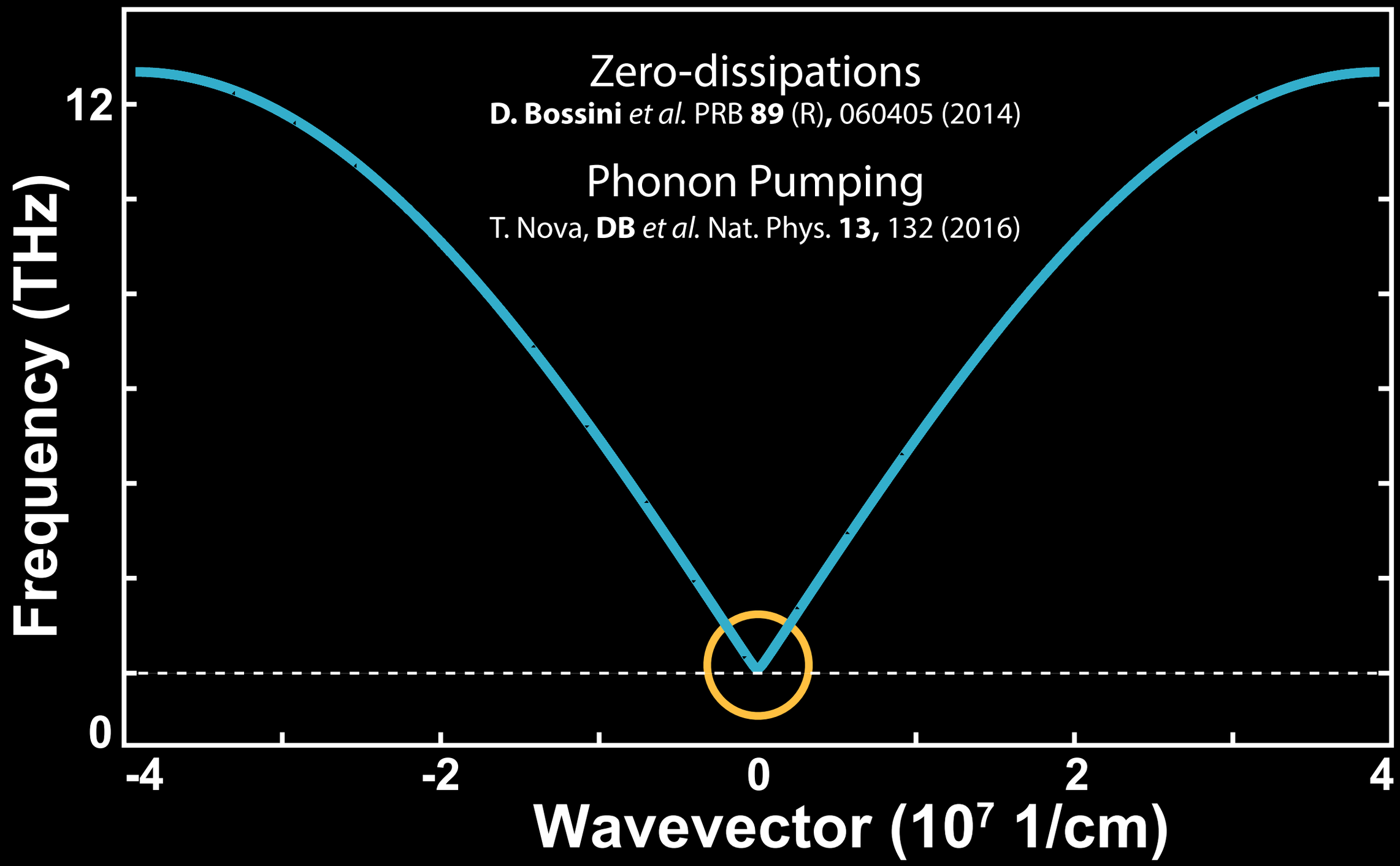


P. Fleury *et al.* Phys. Rev. **2,** 514 (1968)

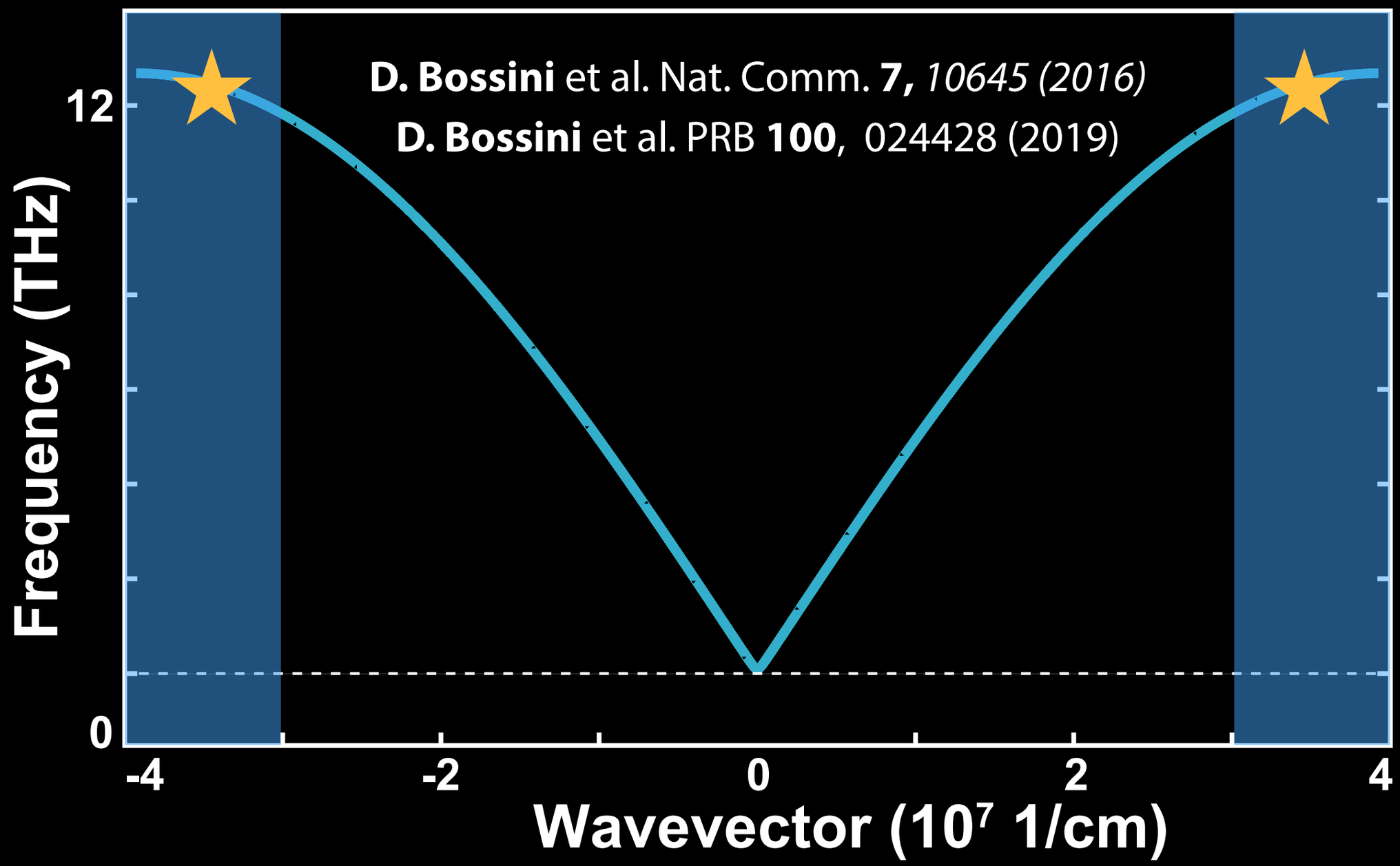


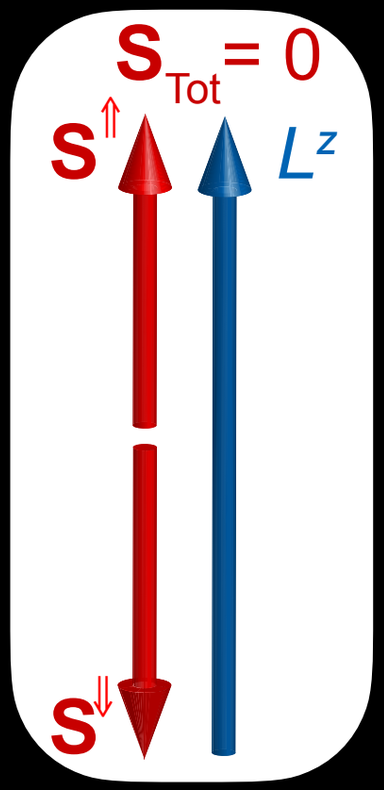
Spin-flip driven by L-S coupling in the excited state

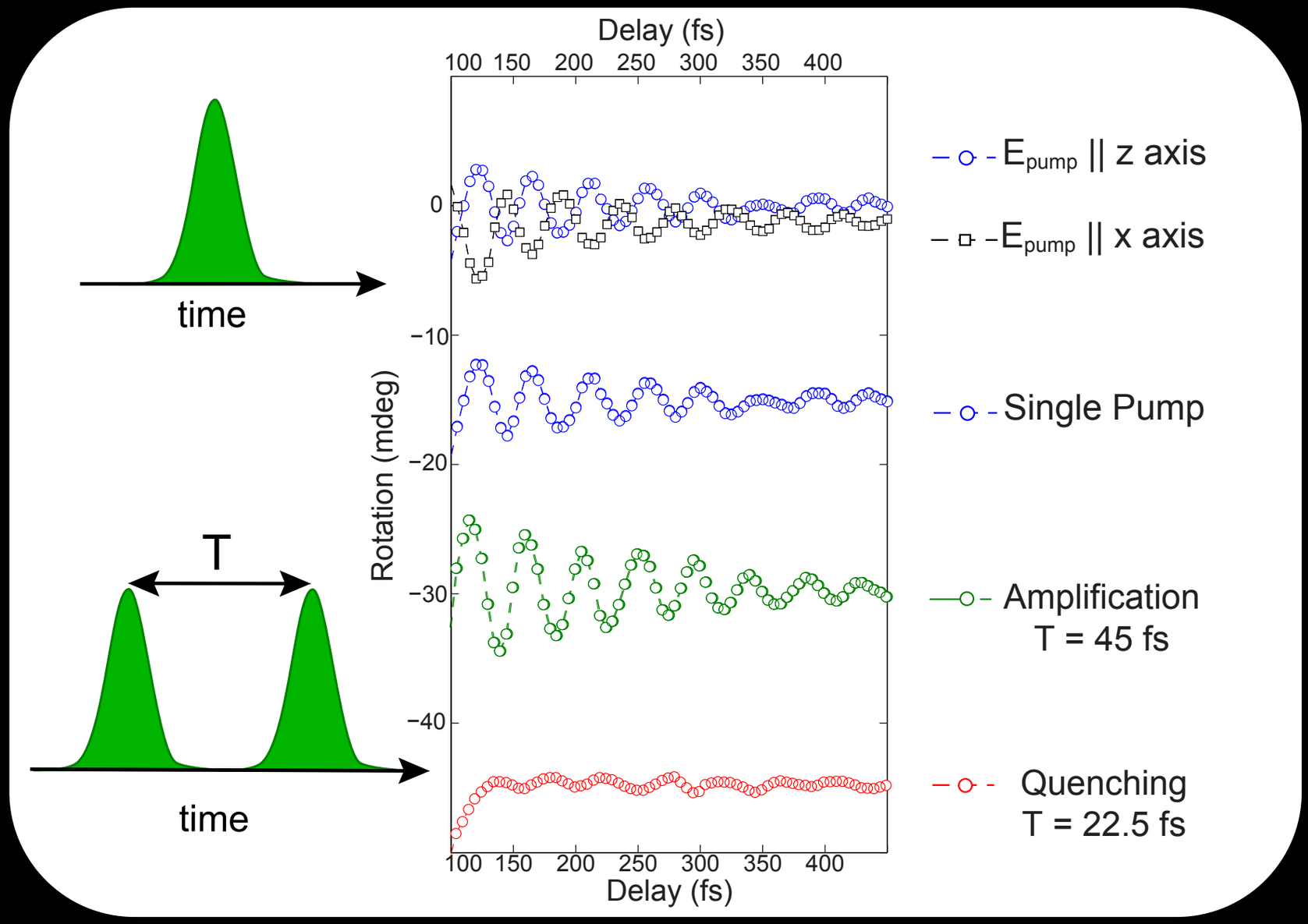
Magnon dispersion



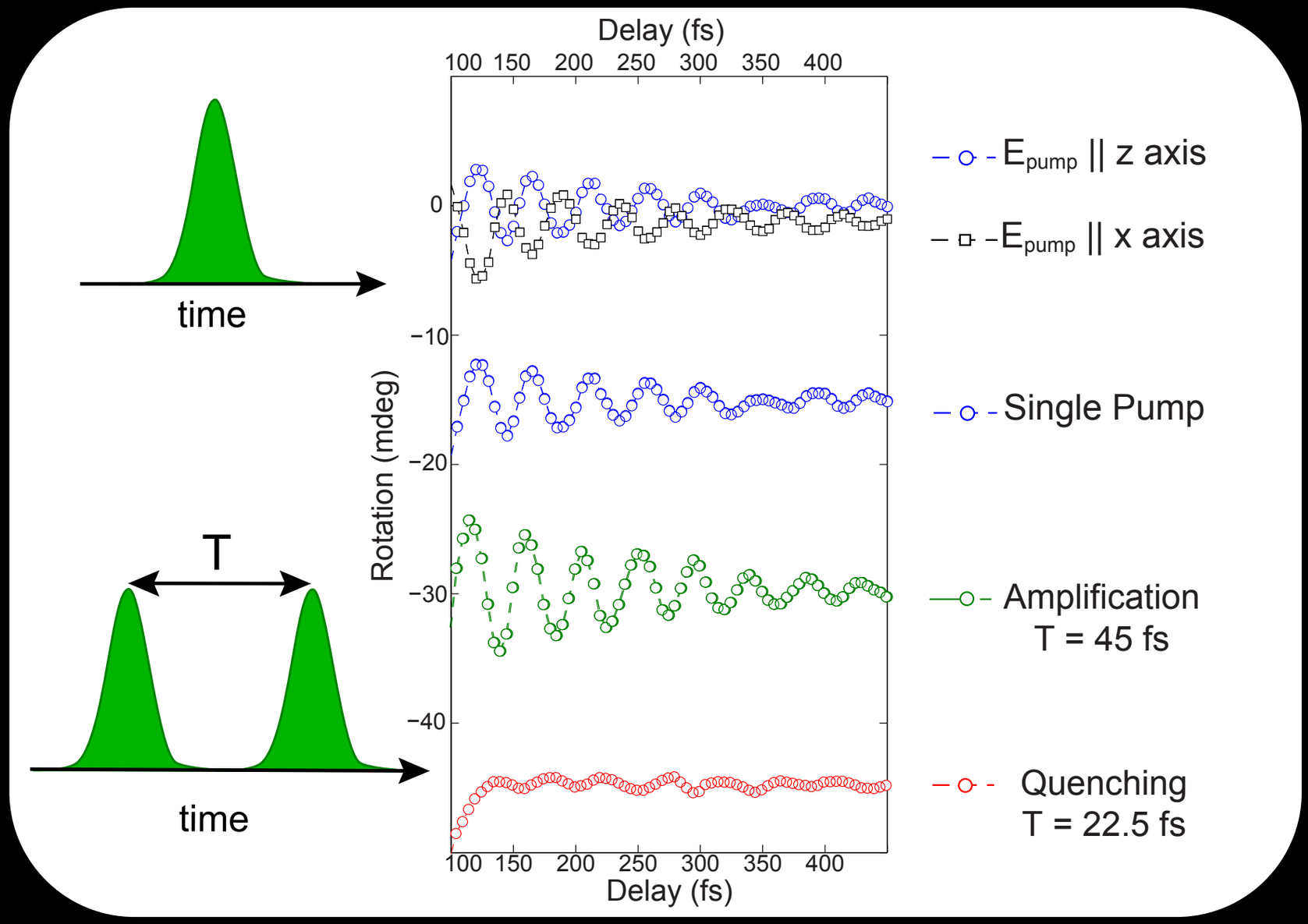
Two-magnon mode





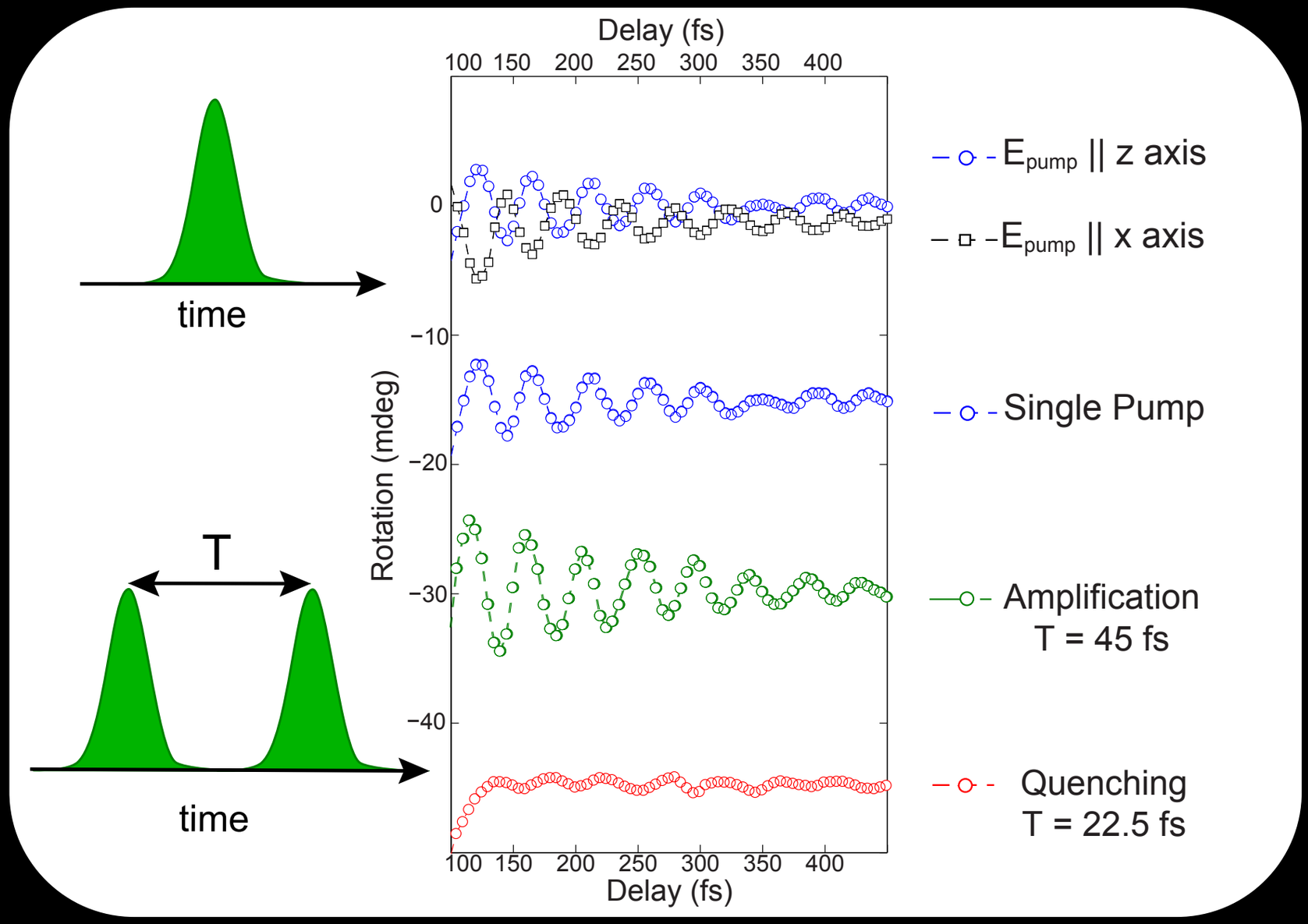


D. Bossini et al. Nat. Comm. **7,** *10645 (2016)*
D. Bossini et al. PRB **100,** *024428 (2019)*



D. Bossini et al. Nat. Comm. **7,** *10645 (2016)*
D. Bossini et al. PRB **100,** *024428 (2019)*

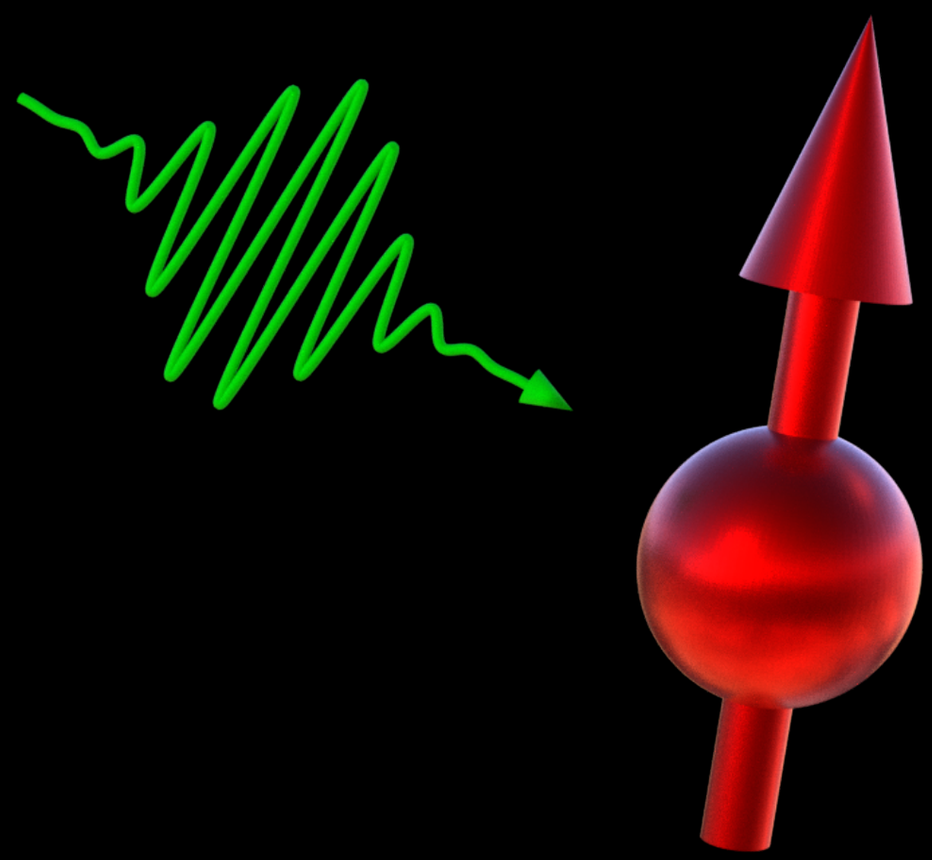
Quantum magnonics @ extreme scales ?



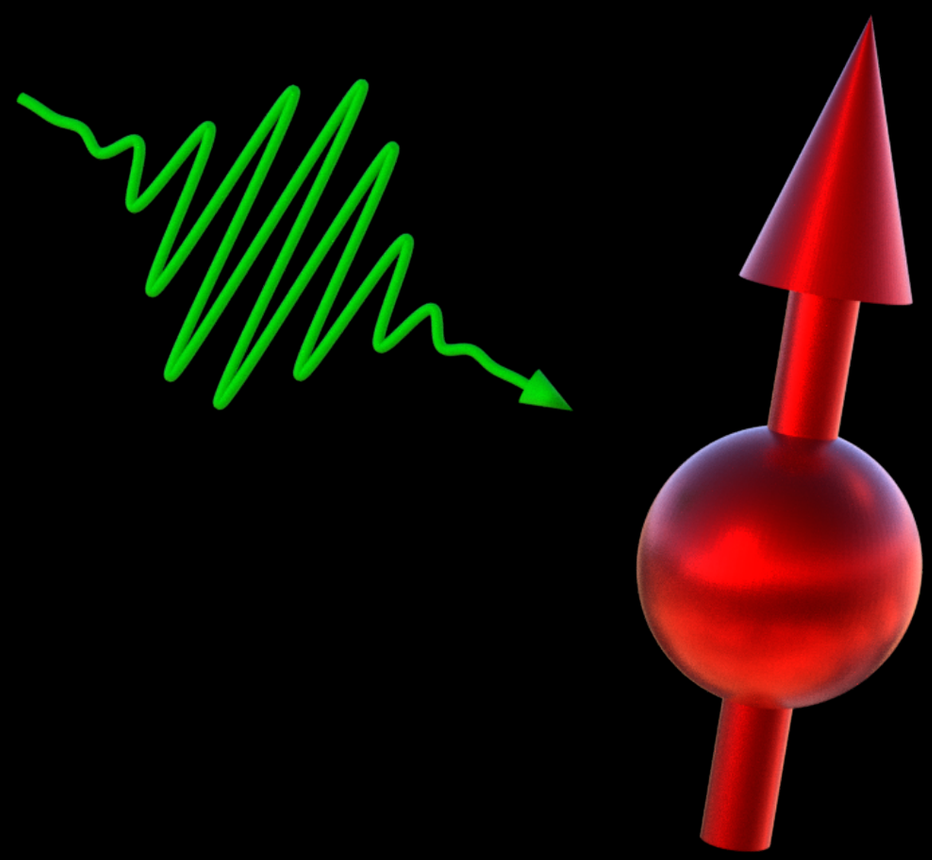
D. Bossini et al. Nat. Comm. **7,** *10645 (2016)*
D. Bossini et al. PRB **100,** *024428 (2019)*

Quantum magnonics @ extreme scales ?
Resonant Pumping??

Open issues



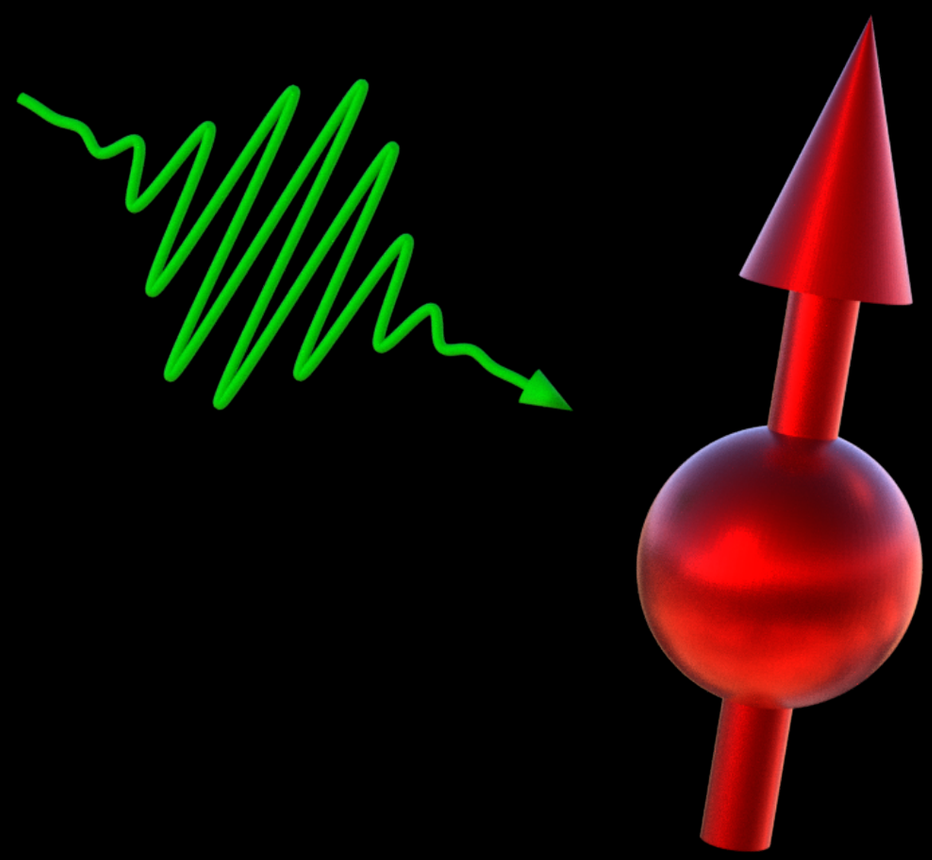
Open issues



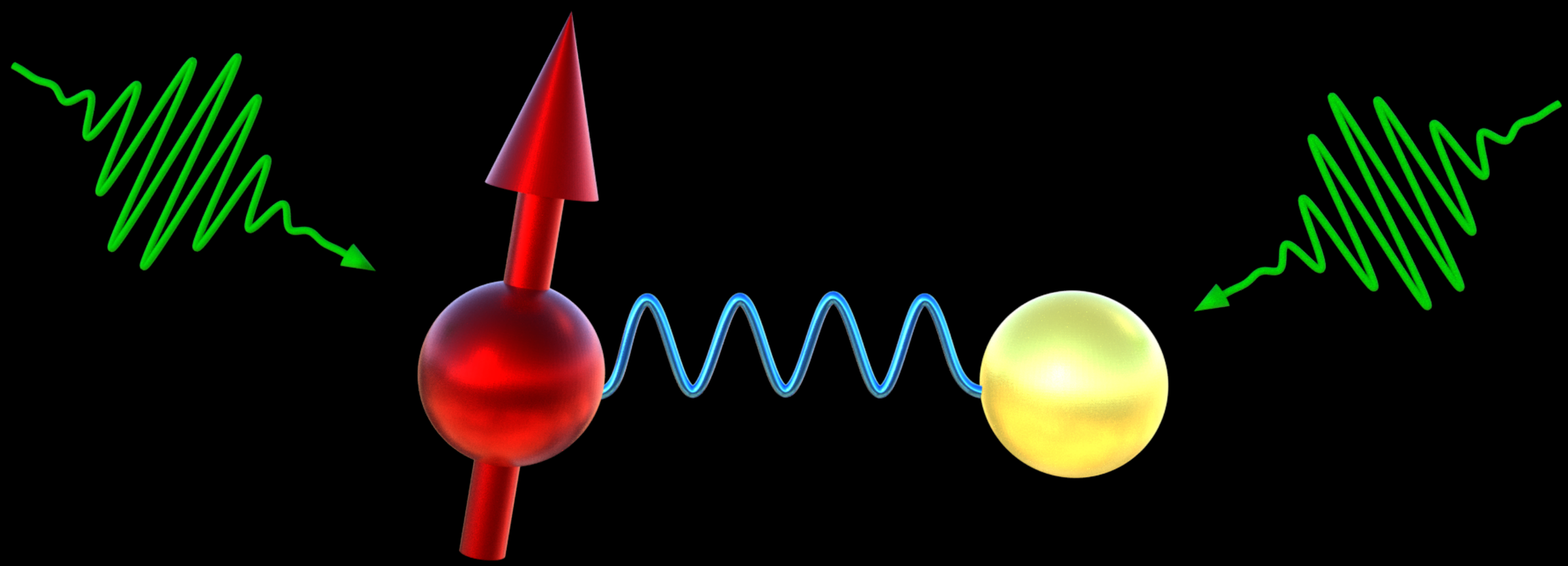
Multi-domain

- Magnon generation and propagation in multi-domain states?

Open issues



Open issues



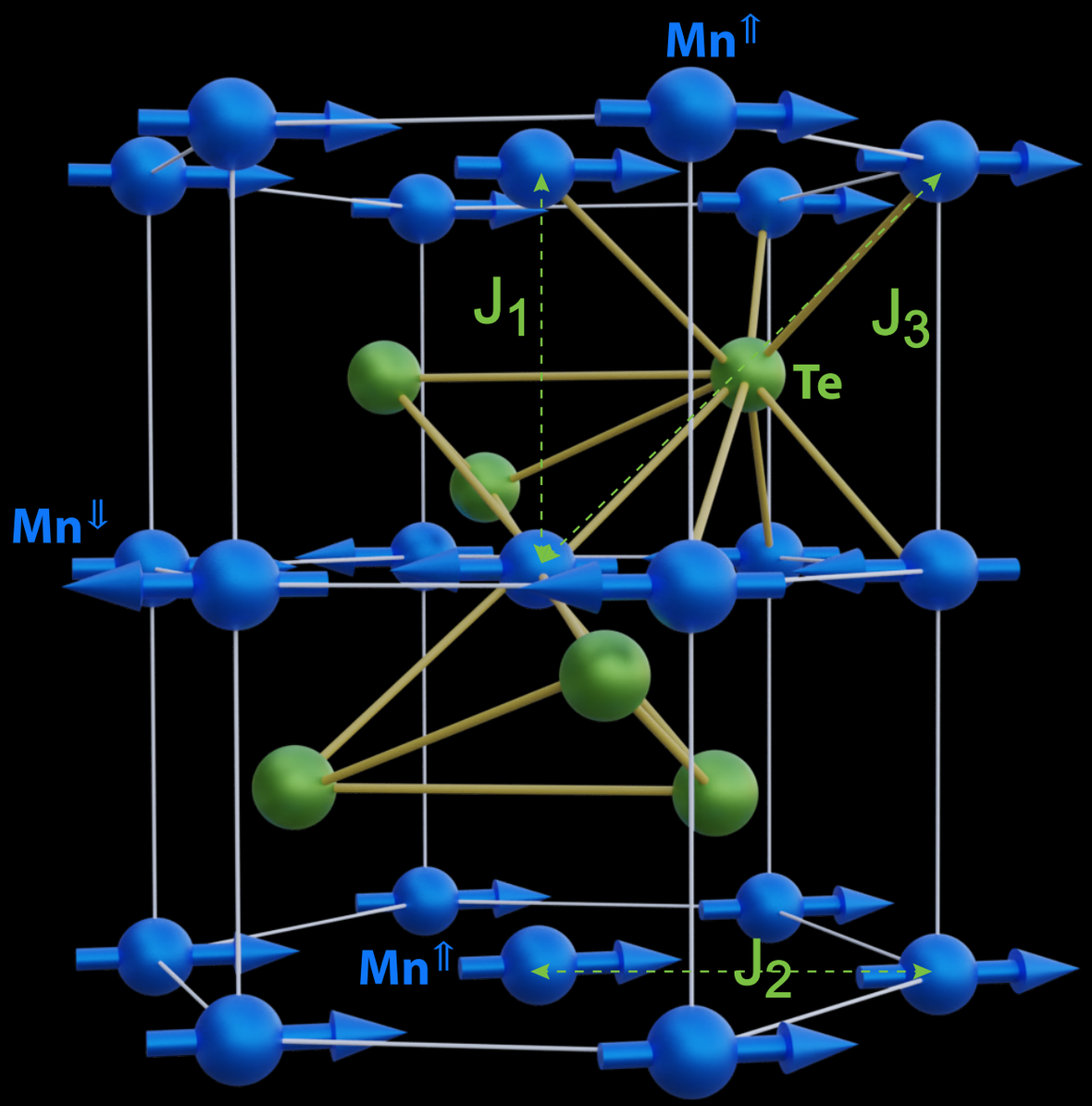
Coupling to charges

- Spin-to-charge coherence/energy transfer
- Single systems / heterostructures
- Manipulation of the coupling

Hexagonal (α)-MnTe

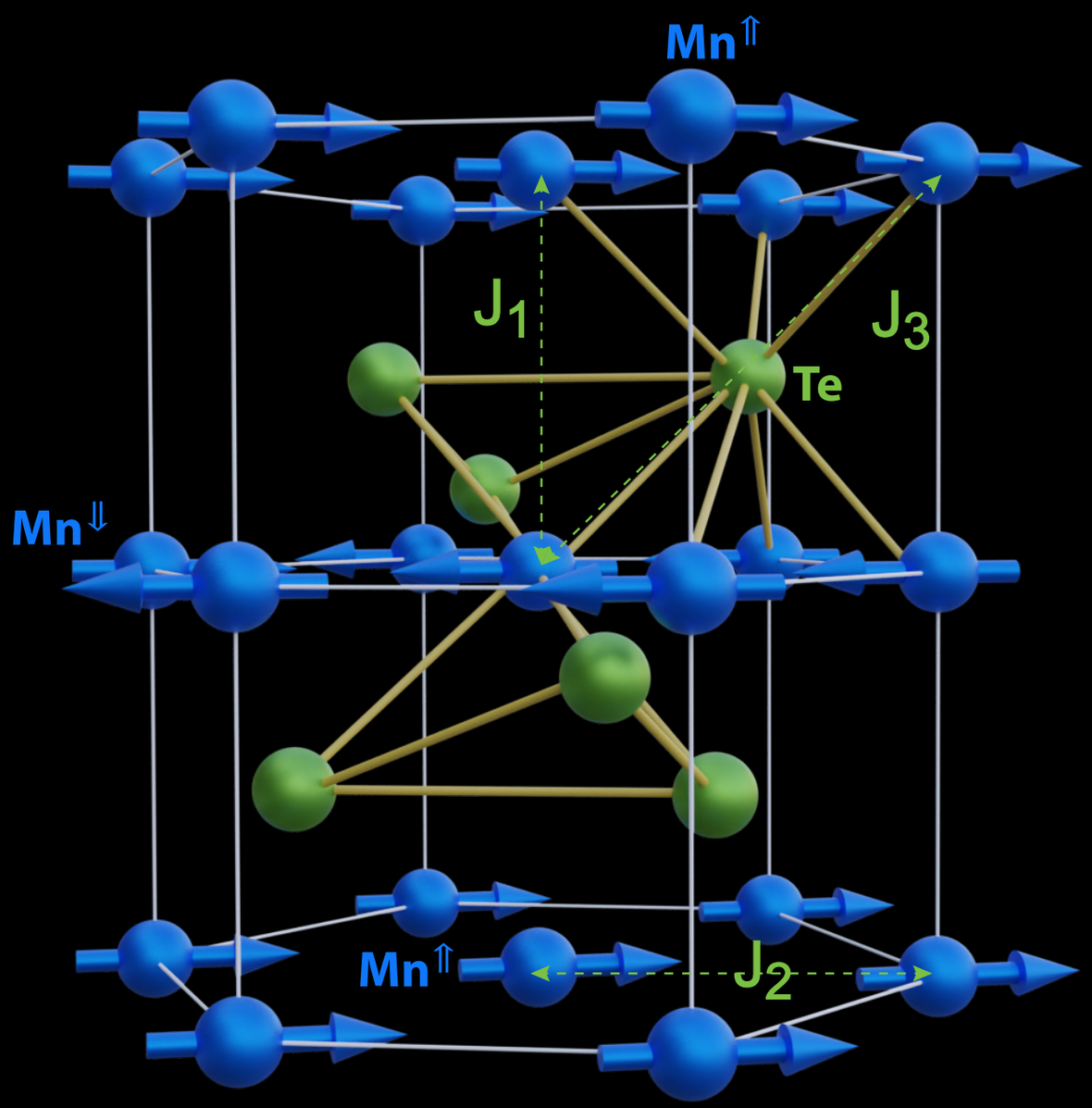
Hexagonal (α)-MnTe

antiferromagnetic
semiconductor



Hexagonal (α)-MnTe

antiferromagnetic
semiconductor

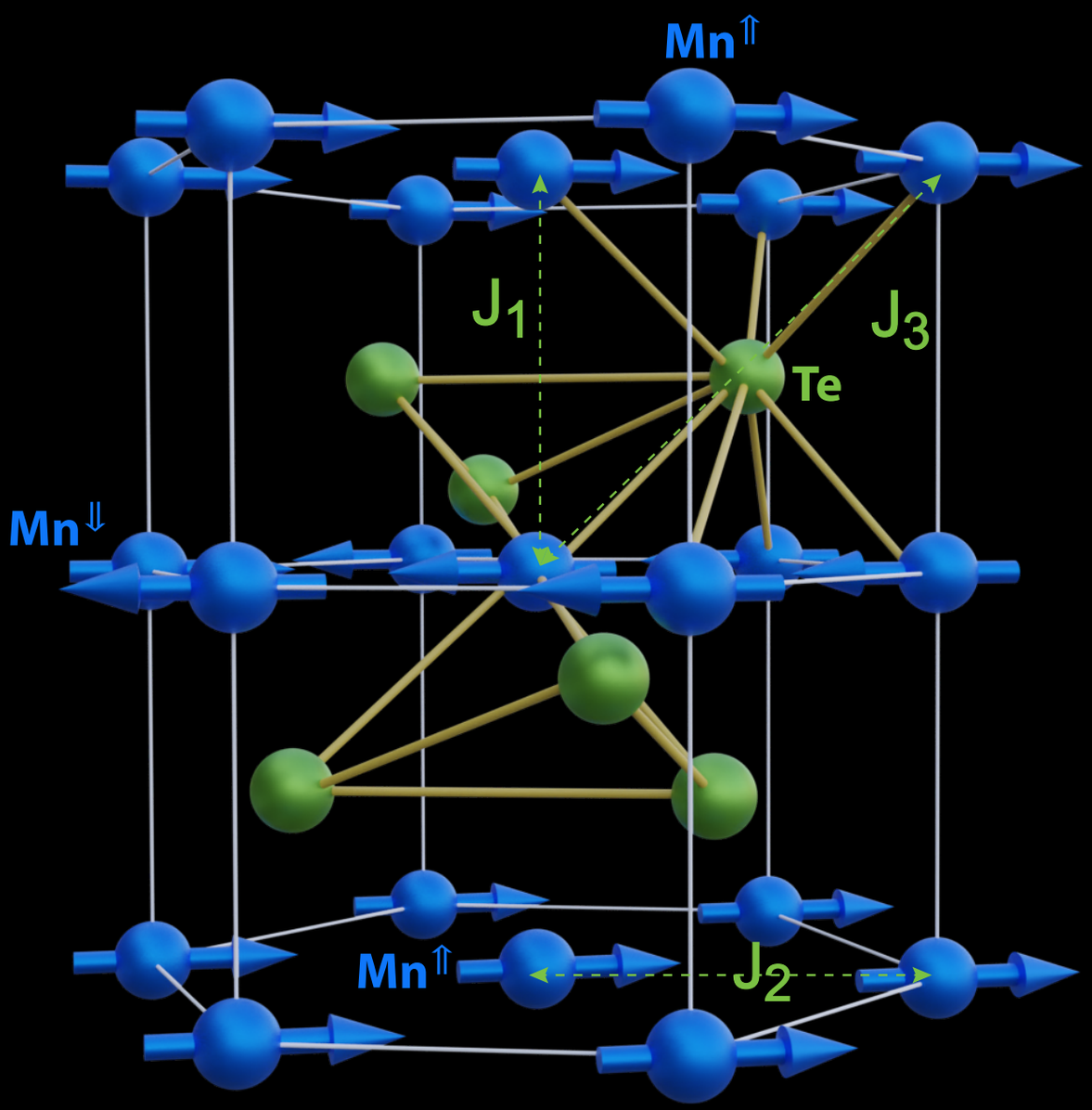


optics magnetism

C. Ferrer-Roca et al. PRB **61**, 13679 (2000)

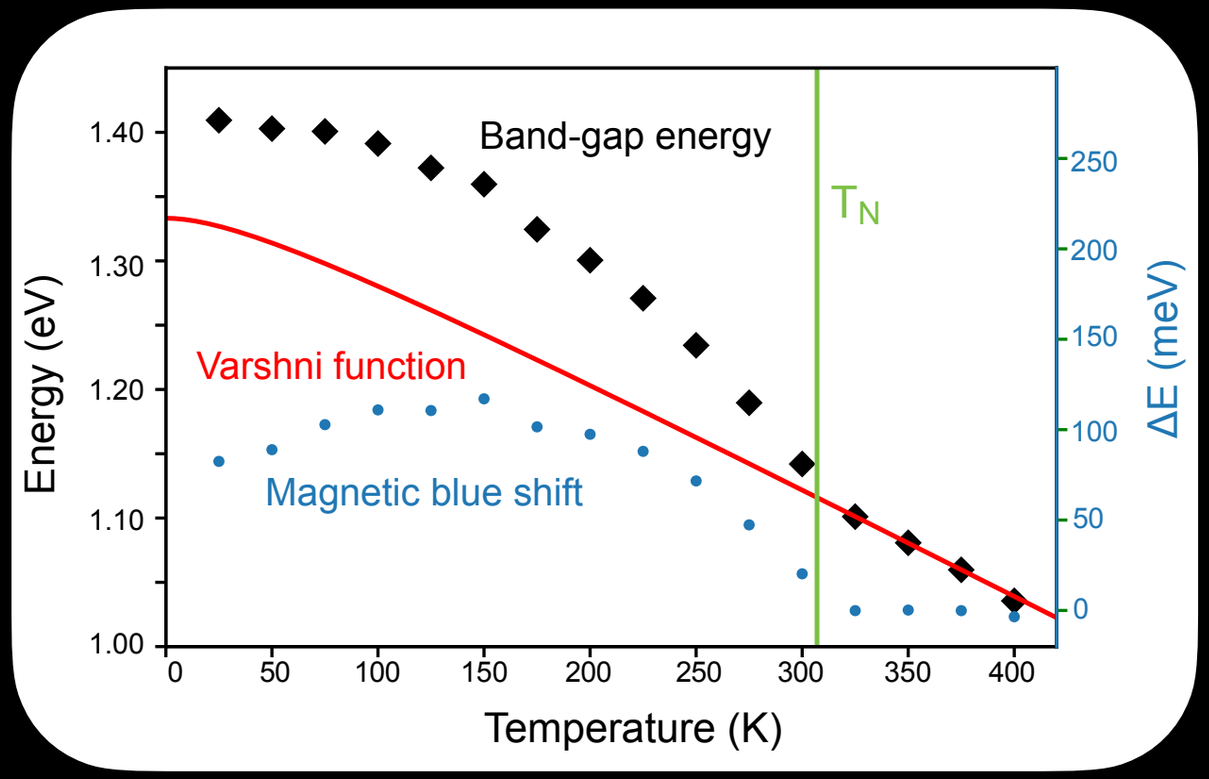
Hexagonal (α)-MnTe

antiferromagnetic semiconductor



optics magnetism

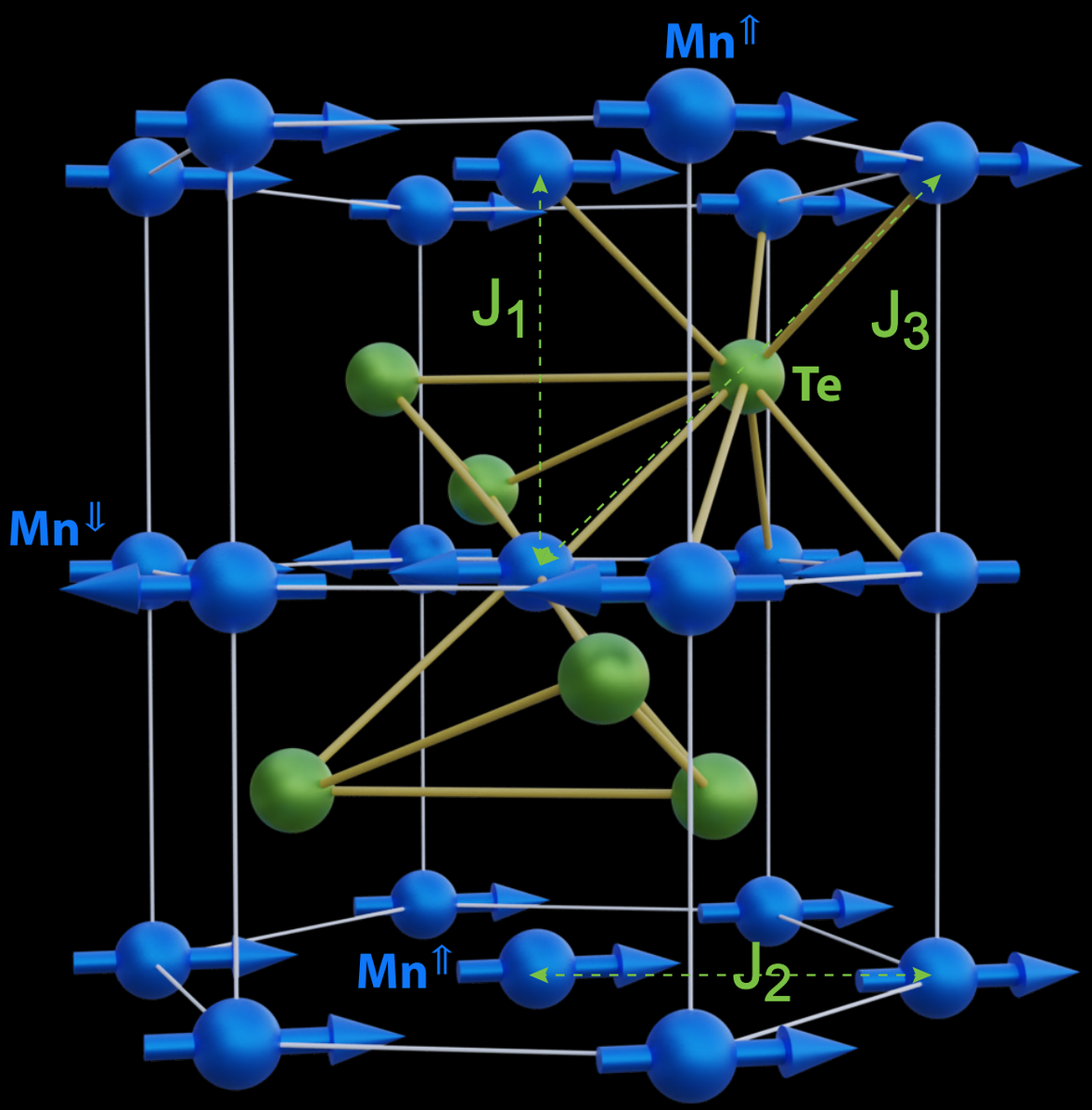
C. Ferrer-Roca et al. PRB **61**, 13679 (2000)



D. Bossini et al. *New J. Phys.* **22**, 083029 (2020)

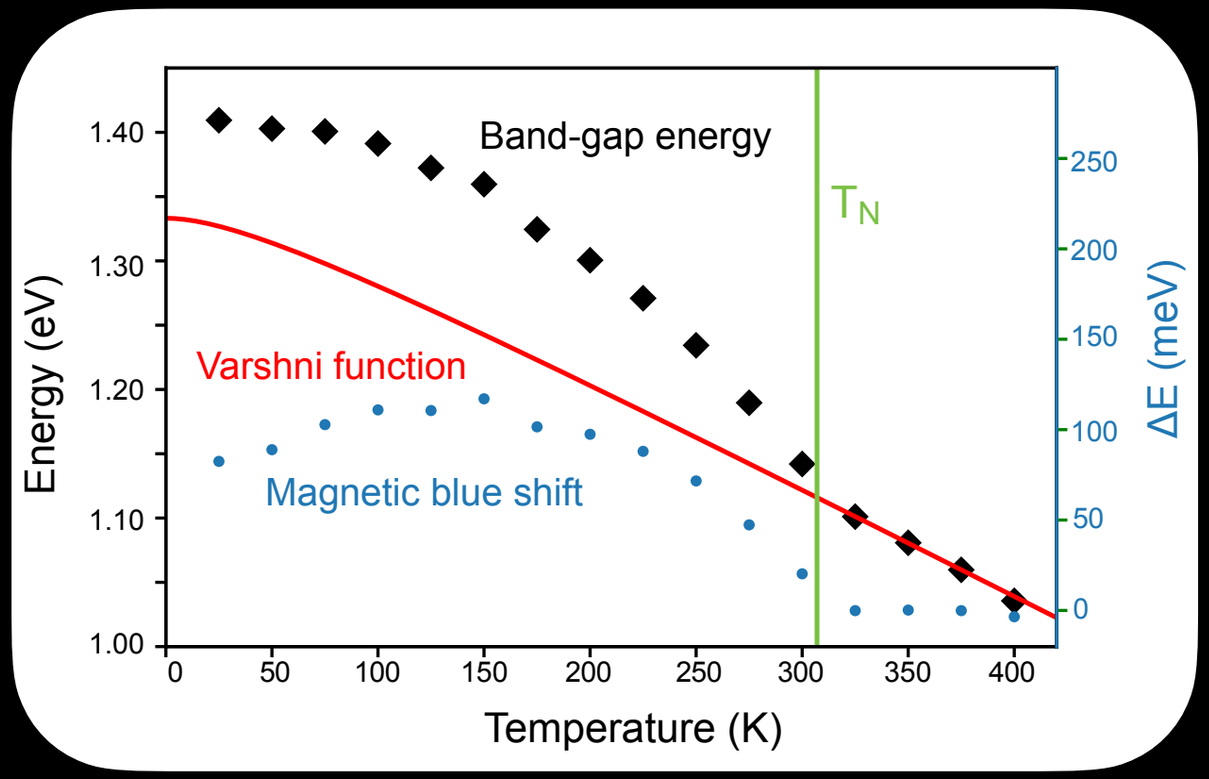
Hexagonal (α)-MnTe

antiferromagnetic semiconductor



optics magnetism

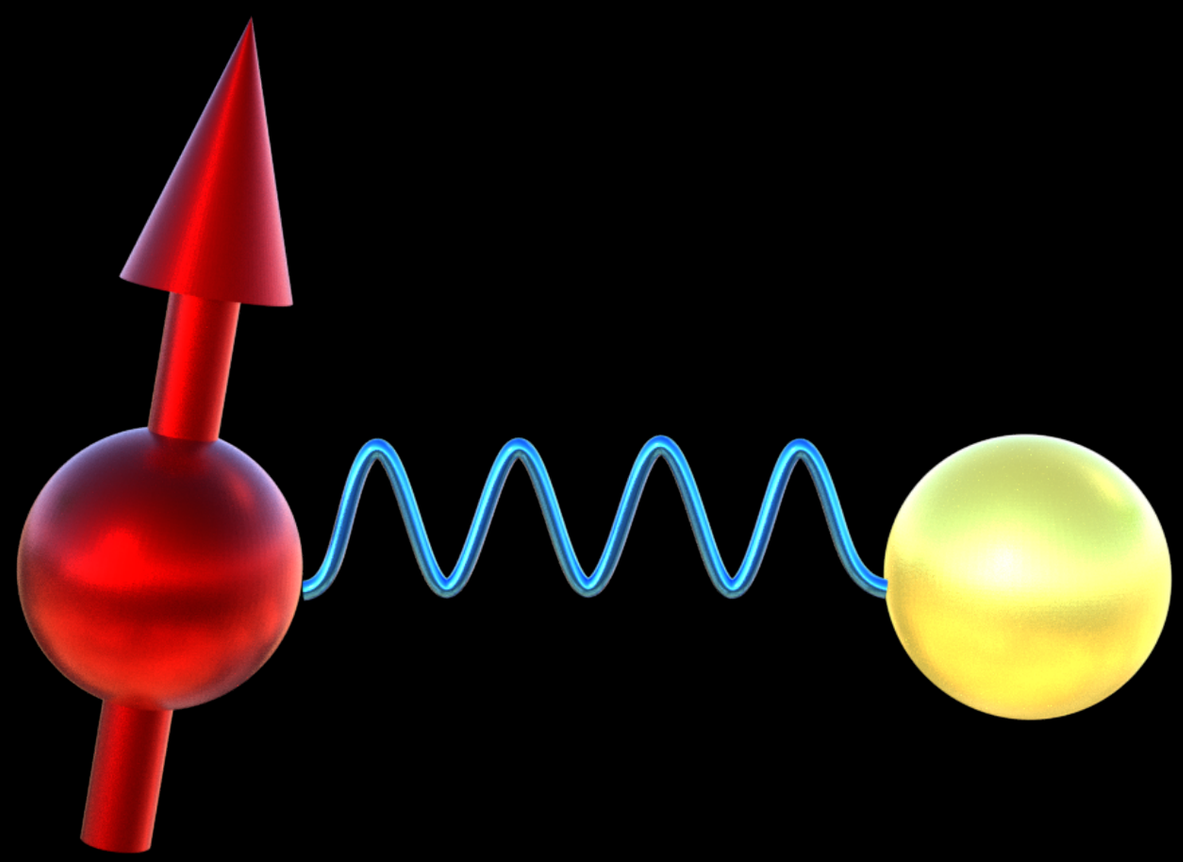
C. Ferrer-Roca et al. PRB **61**, 13679 (2000)



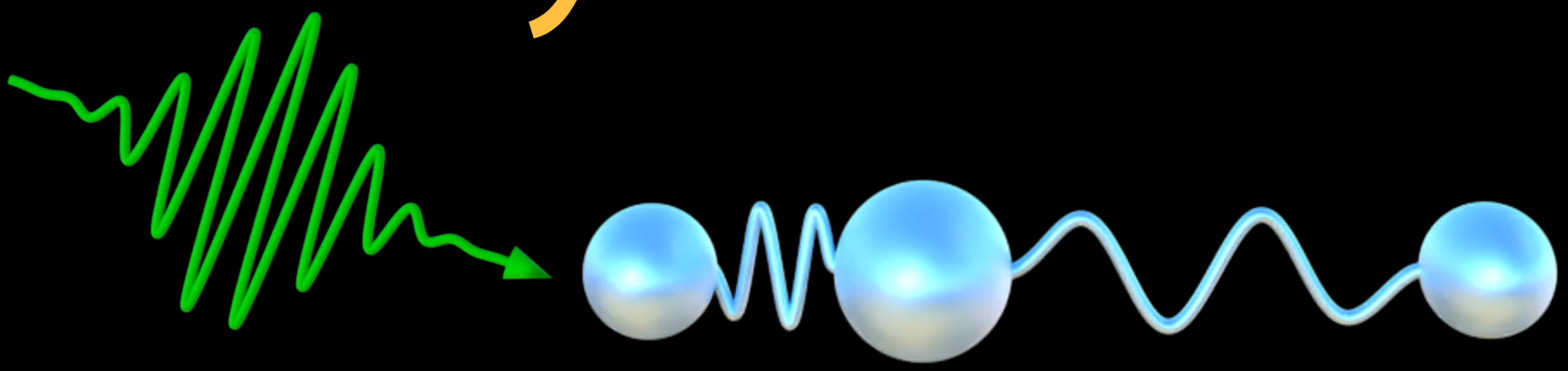
D. Bossini et al. *New J. Phys.* **22**, 083029 (2020)

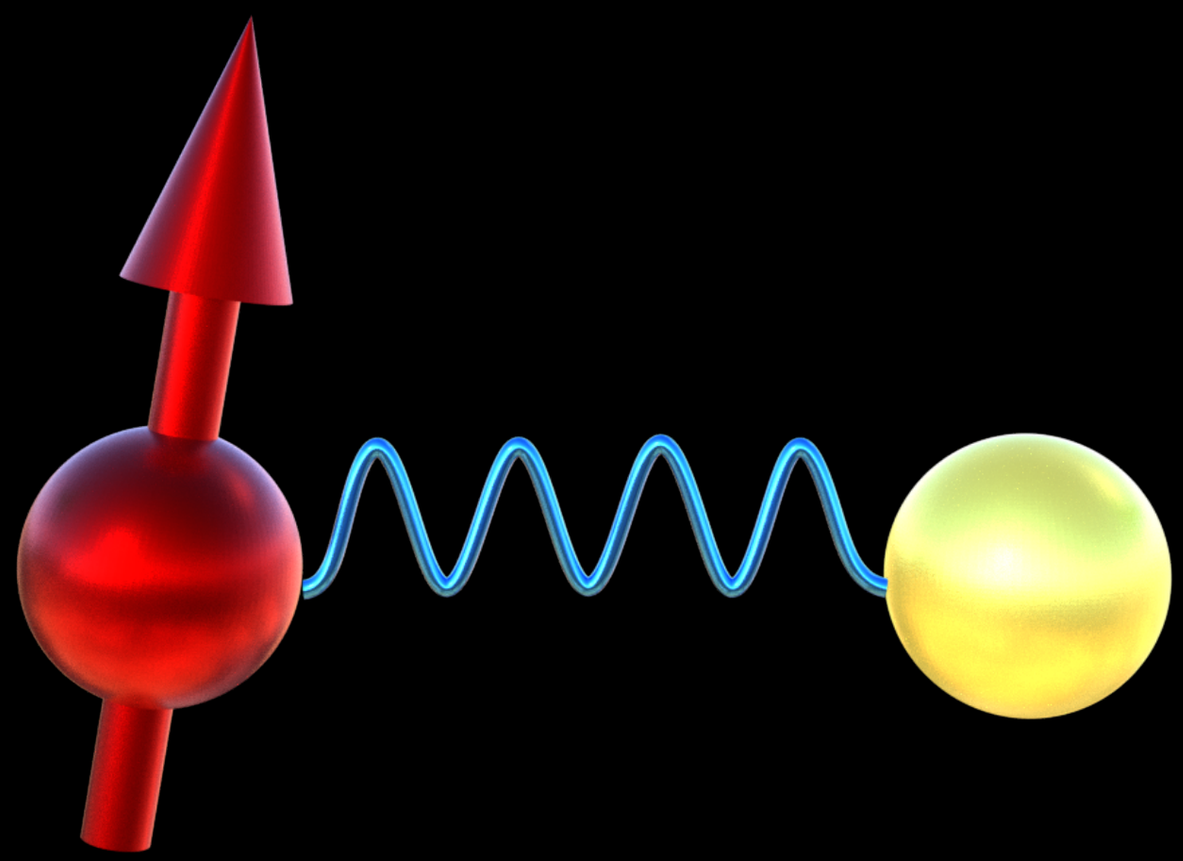
M. Thorbati, **DB** et al. *Phys. Rev. Res. 3, 043232 (2021)*

Dynamics in MnTe

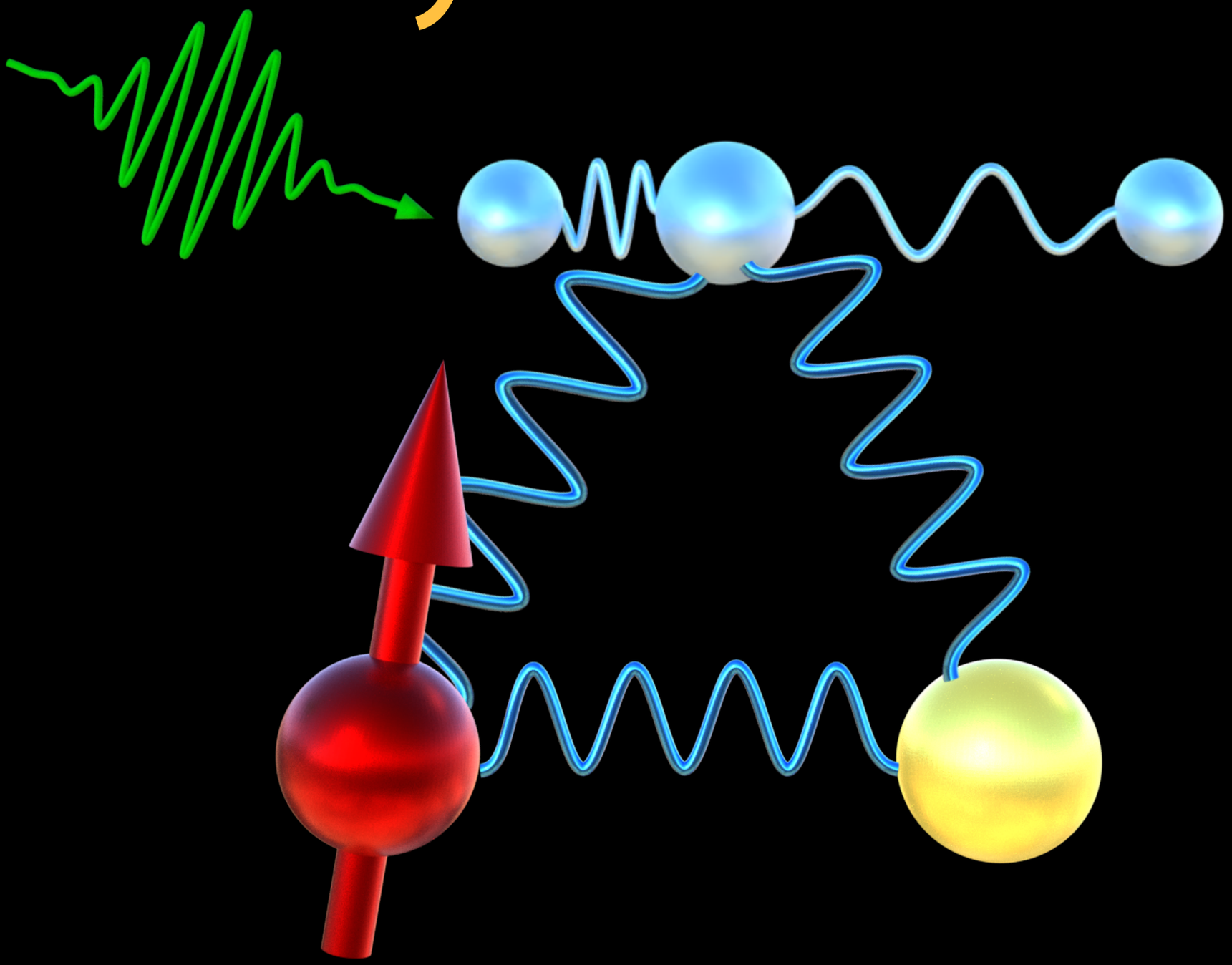


Dynamics in MnTe

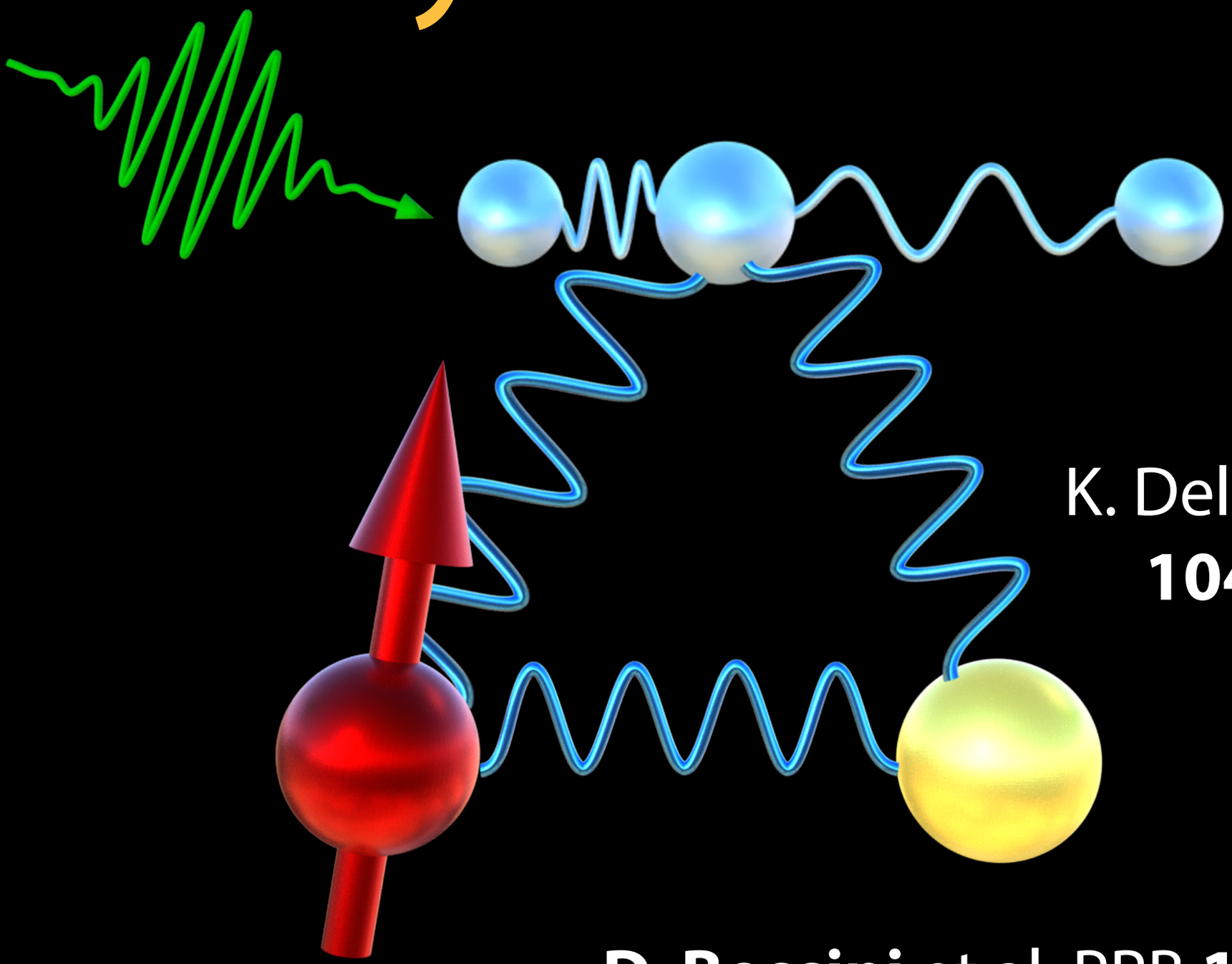




Dynamics in MnTe



Dynamics in MnTe

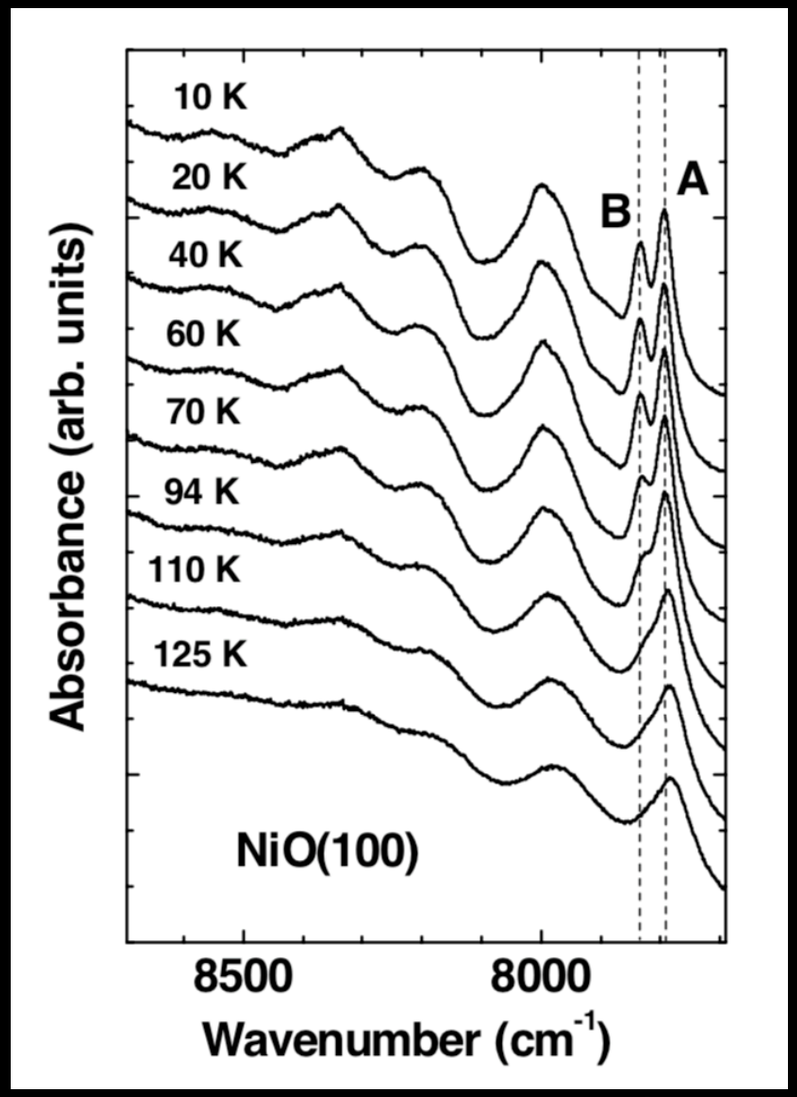


K. Deltenre, **DB** et al. PRB **104**, 184419 (2021)

D. Bossini et al. PRB **104**, 224424 (2021)

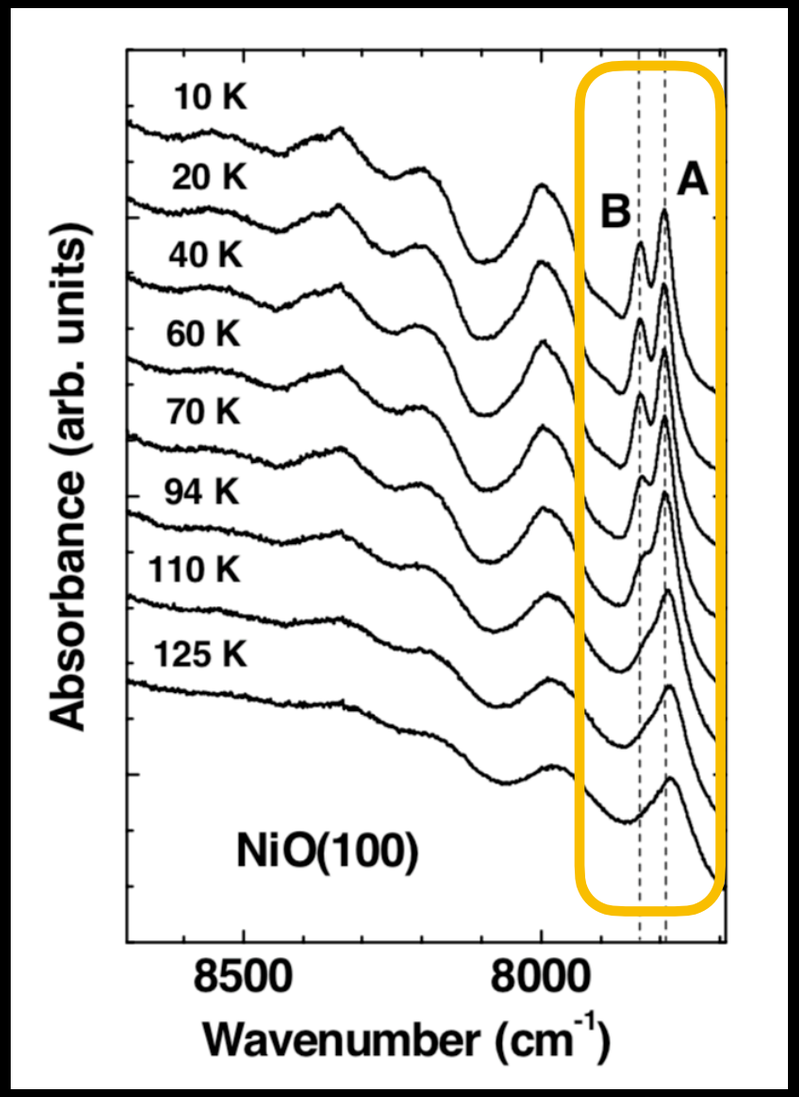
Exciton-magnon transition

Exciton-magnon transition



N. Mironova-Ulmane et al. Proceedings of SPIE 5946 (2012)

Exciton-magnon transition



N. Mironova-Ulmane et al. Proceedings of SPIE 5946 (2012)

- Absorption spectrum AF dielectrics
- Below gap
- Side-band structure
- Spin-forbidden transition

X-M mechanism

Moriya et al. PRL **15**, 1023 (1965) Tanabe et al. *Excitons in Magnetic Insulators* (1982)

Spin-forbidden transition + Magnon

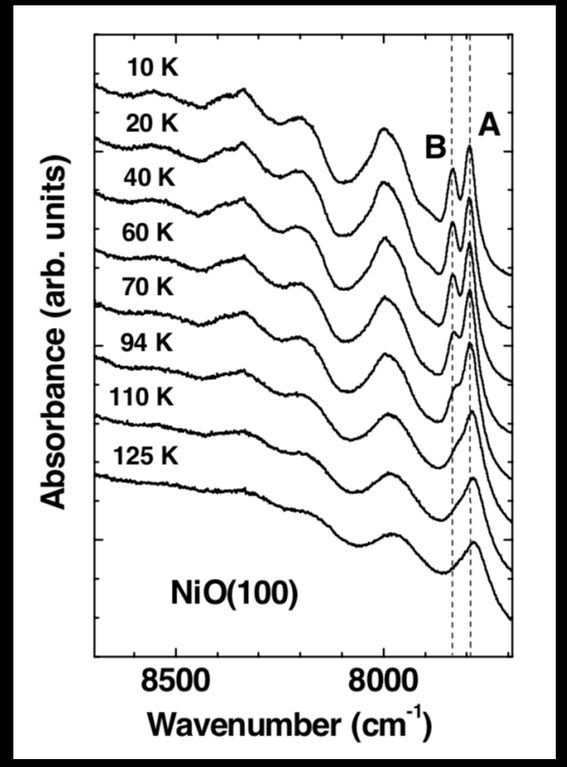
$\Delta S = 1$ $\Delta S = -1$

X-M mechanism

Moriya et al. PRL **15**, 1023 (1965) Tanabe et al. *Excitons in Magnetic Insulators* (1982)

Spin-forbidden transition + Magnon

$\Delta S = 1$ $\Delta S = -1$

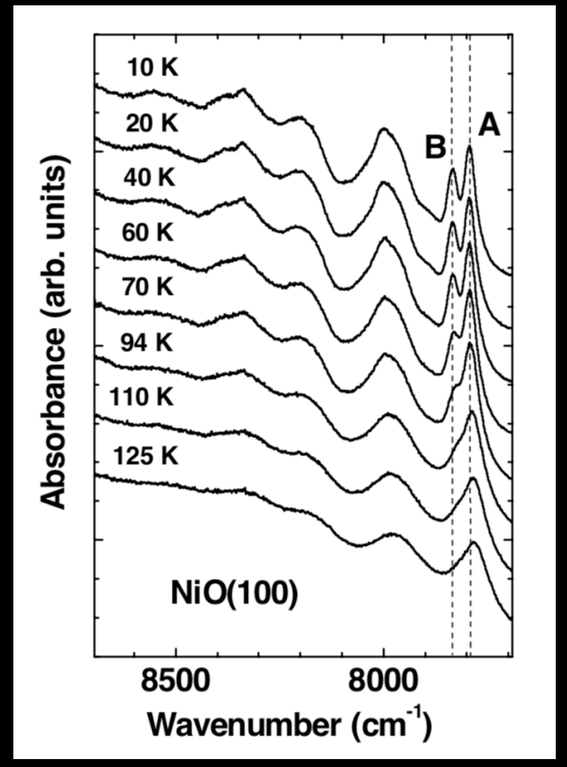


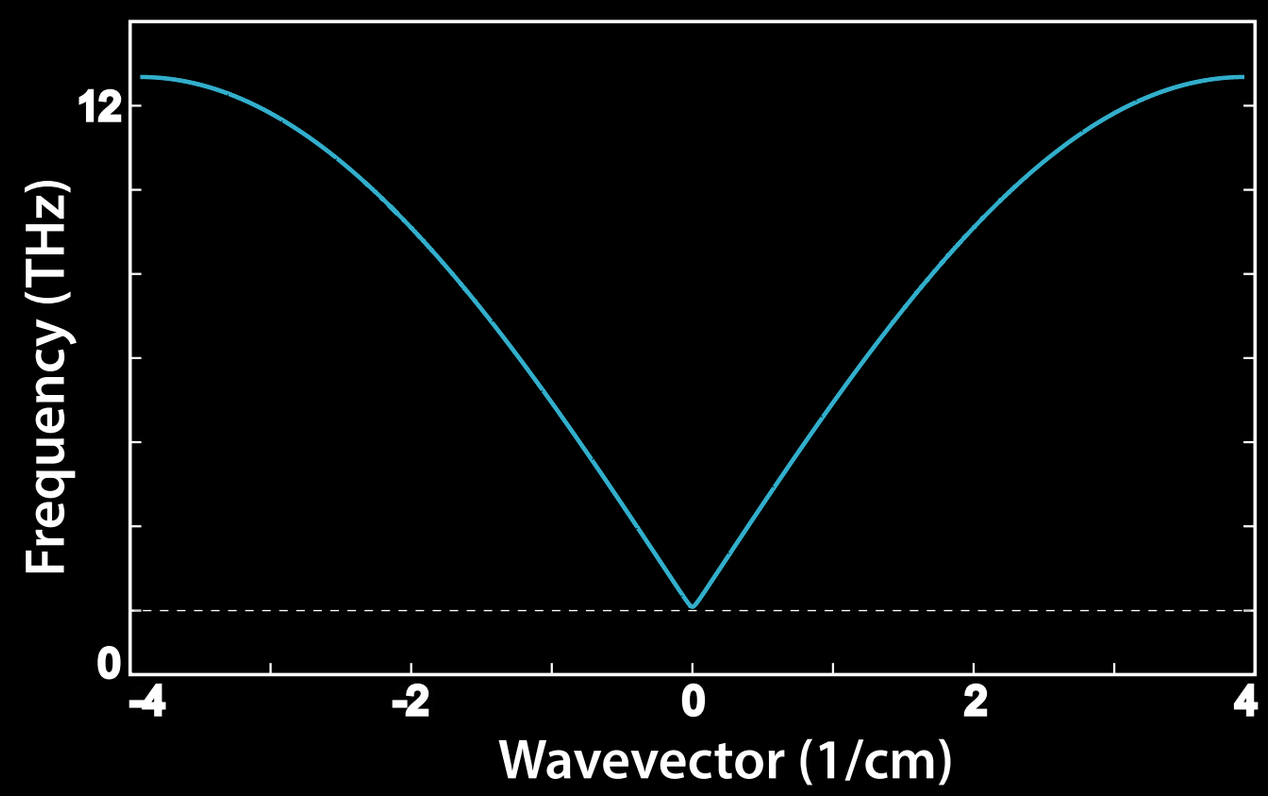
X-M mechanism

Moriya et al. PRL **15**, 1023 (1965) Tanabe et al. *Excitons in Magnetic Insulators* (1982)

Spin-forbidden transition + Magnon

$\Delta S = 1$ $\Delta S = -1$



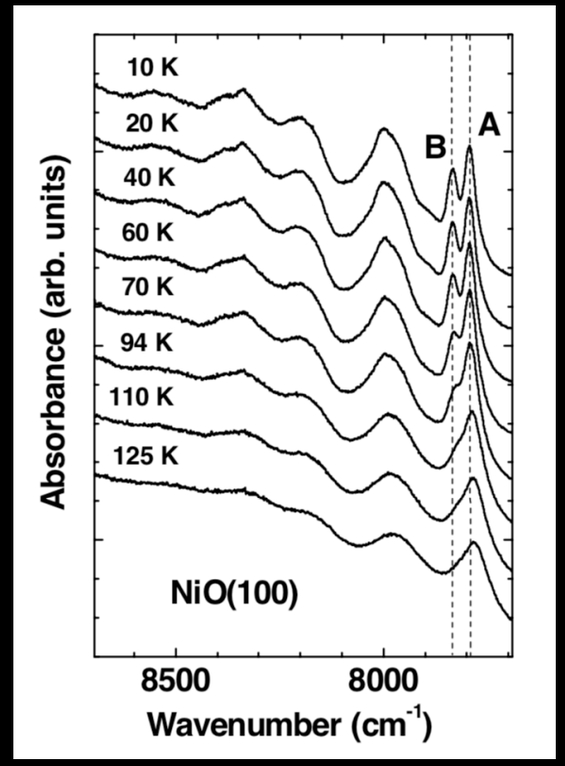


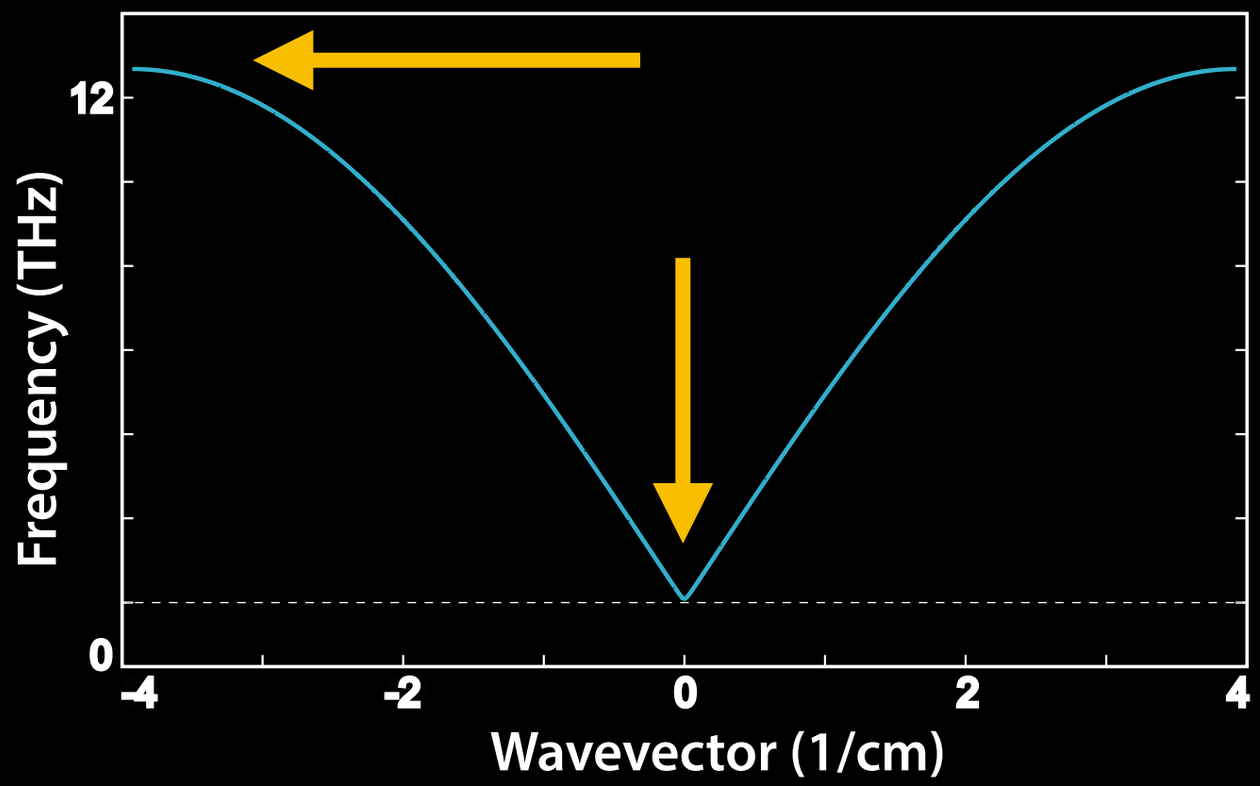
X-M mechanism

Moriya et al. PRL **15**, 1023 (1965) Tanabe et al. *Excitons in Magnetic Insulators* (1982)

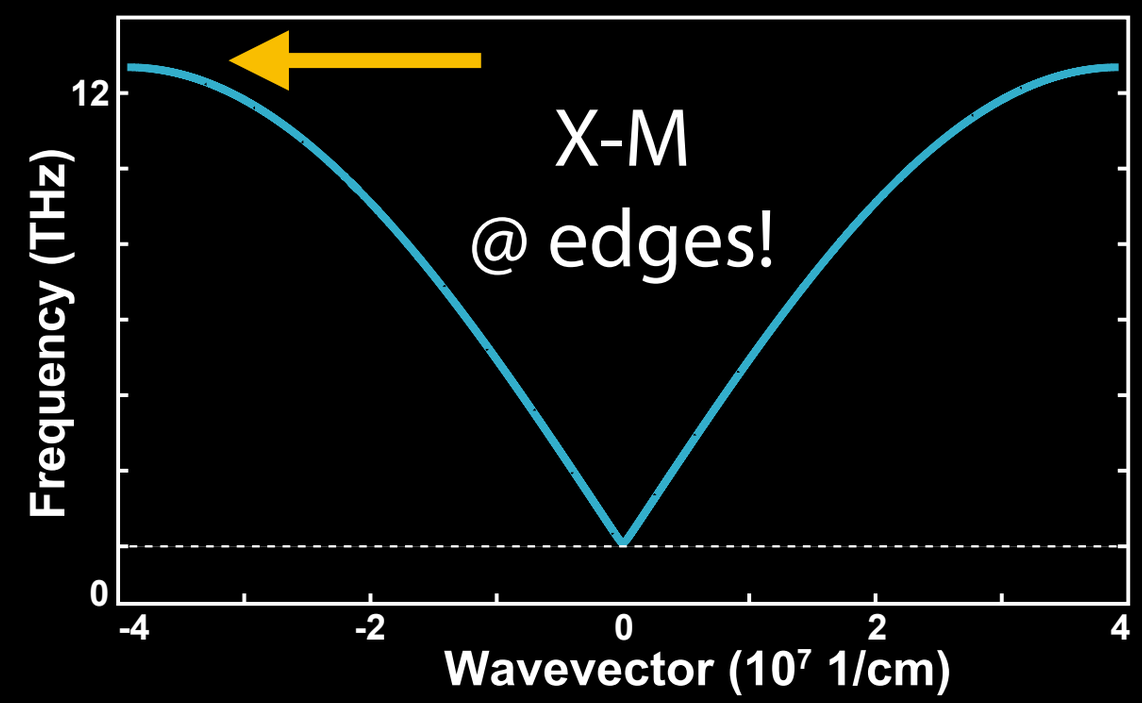
Spin-forbidden transition + Magnon

$\Delta S = 1$ $\Delta S = -1$



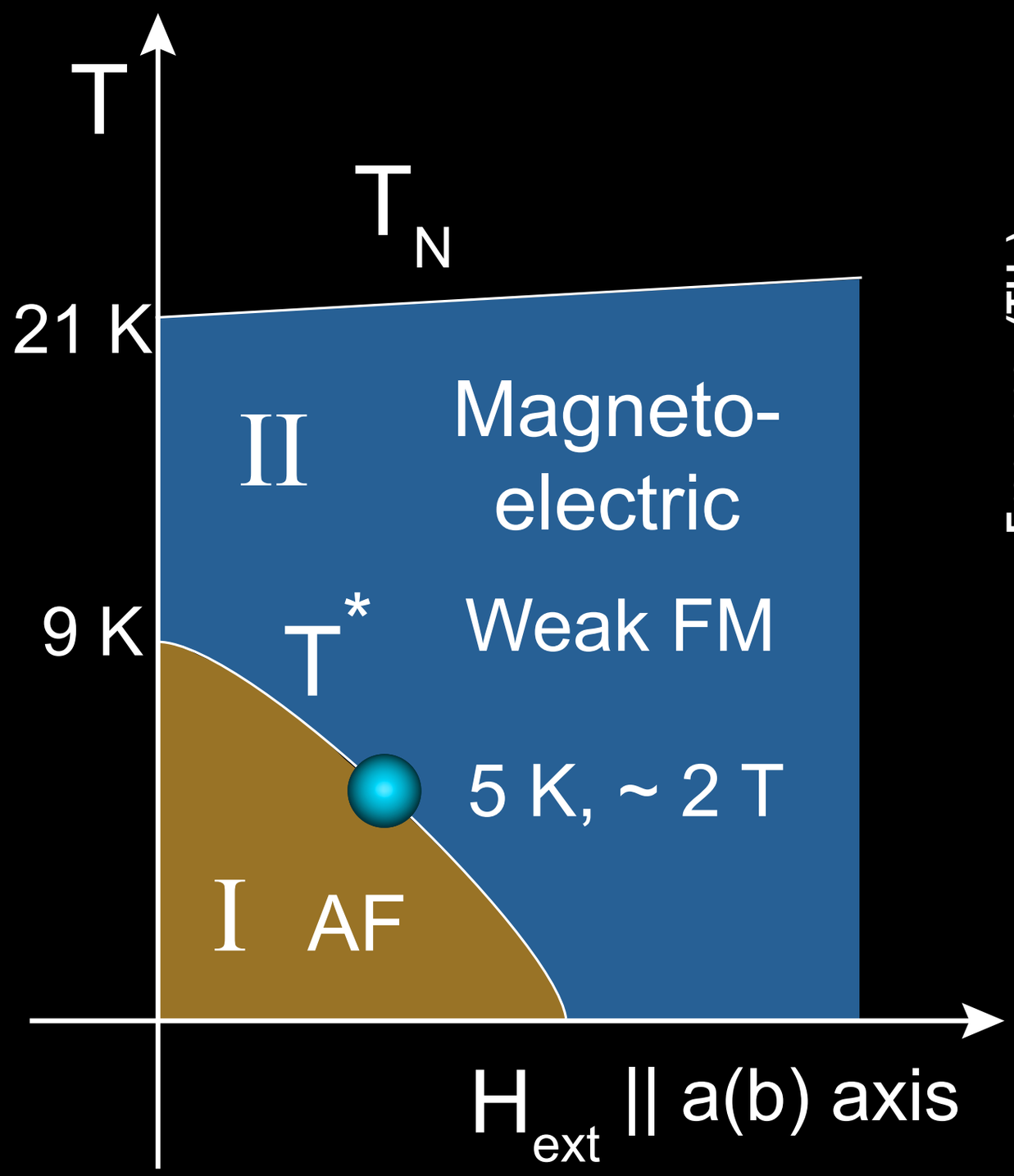


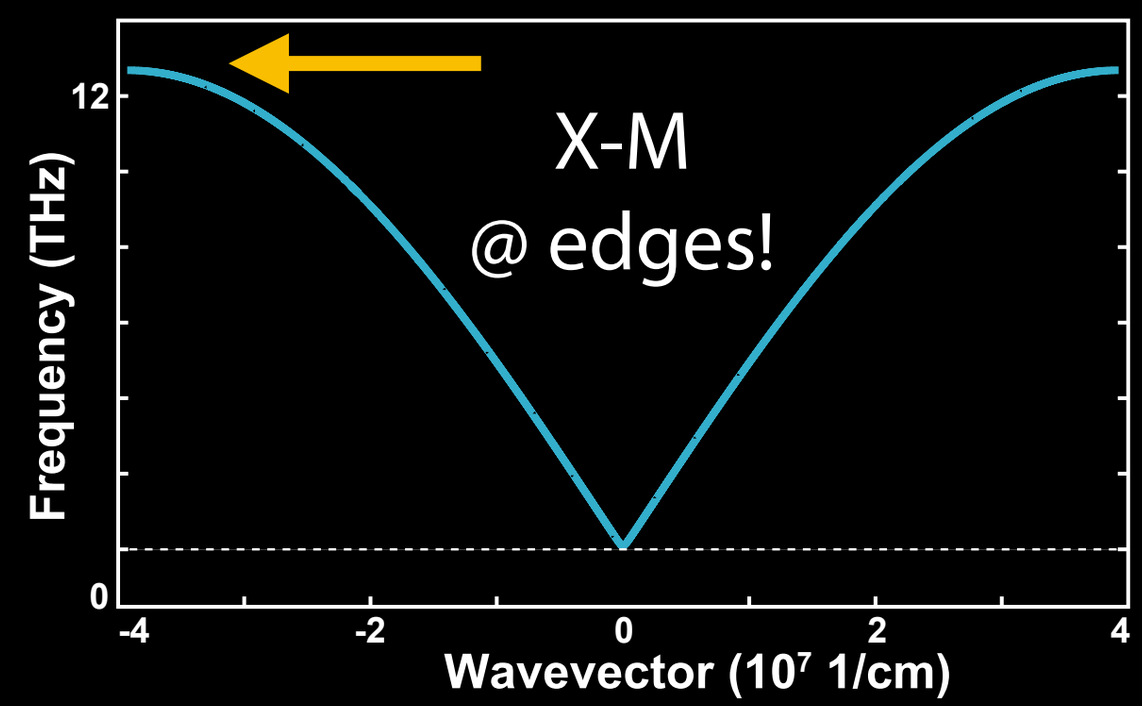
CuB_2O_4 phase diagram



Toyoda et al. PRB. **93**, 201109 (2016)
Martynov et al. J. MMM **299**, 75 (2006)

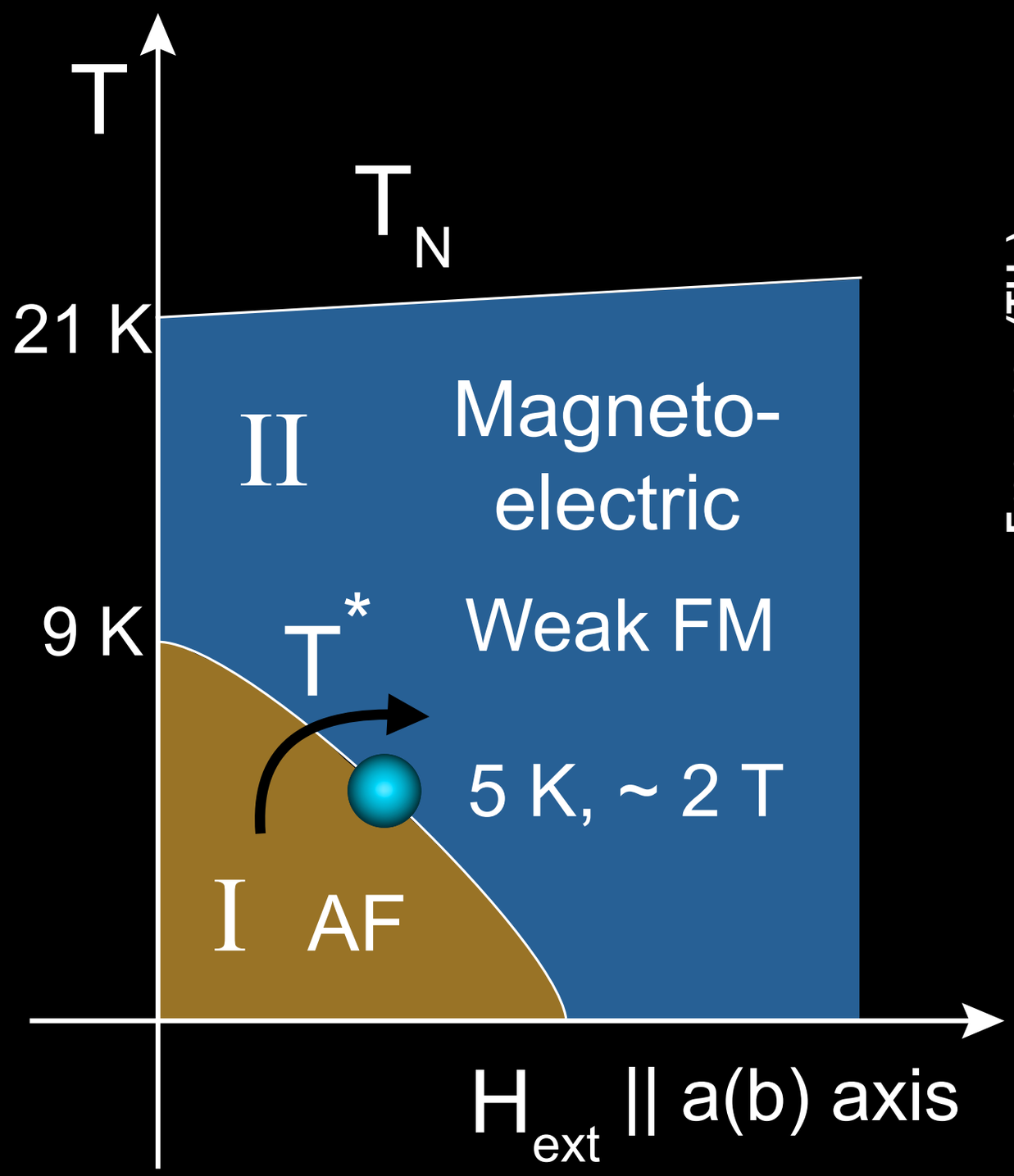
CuB_2O_4 phase diagram

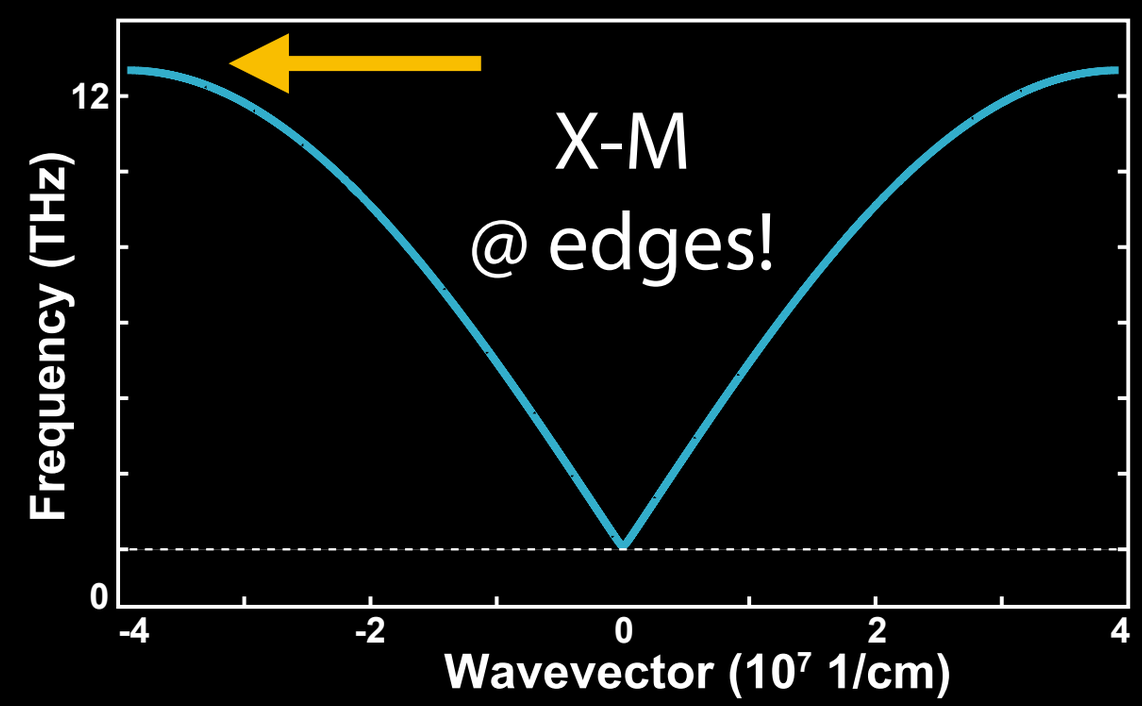




Toyoda et al. PRB. **93**, 201109 (2016)
Martynov et al. J. MMM **299**, 75 (2006)

CuB_2O_4 phase diagram



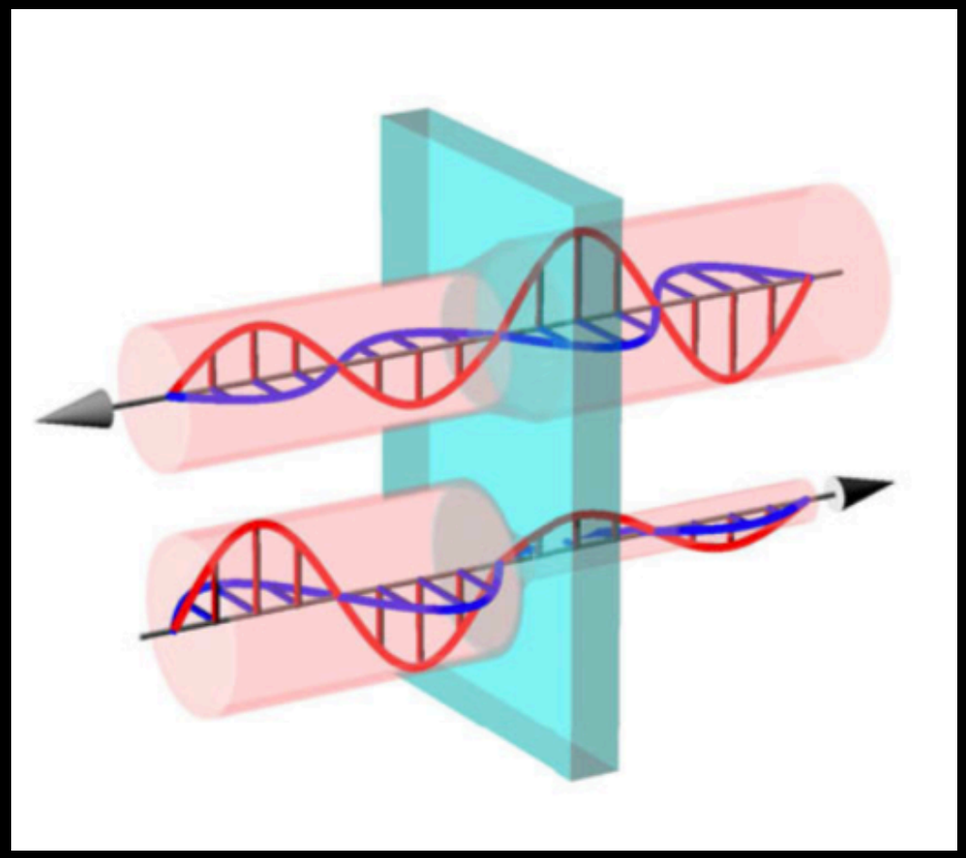


Toyoda et al. PRB. **93**, 201109 (2016)
Martynov et al. J. MMM **299**, 75 (2006)

Phase transition?

Optical ME detection

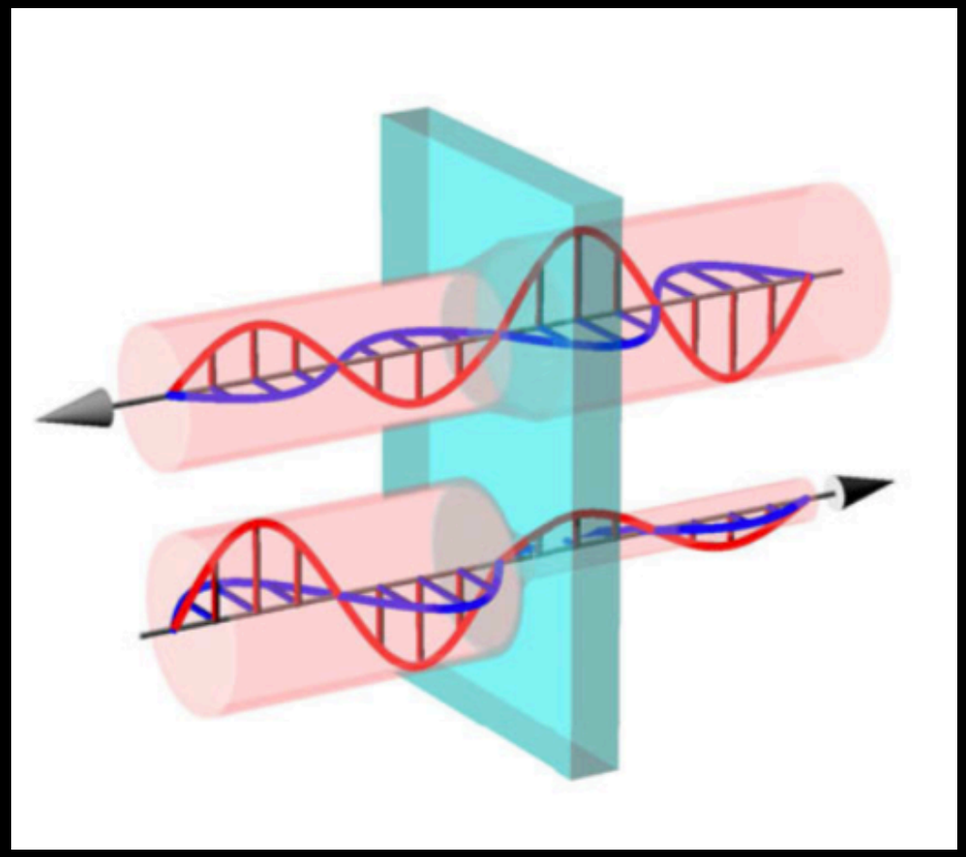
Optical ME detection



S. Toyoda et al., Phys. Rev. Lett 115, 267207 (2015)

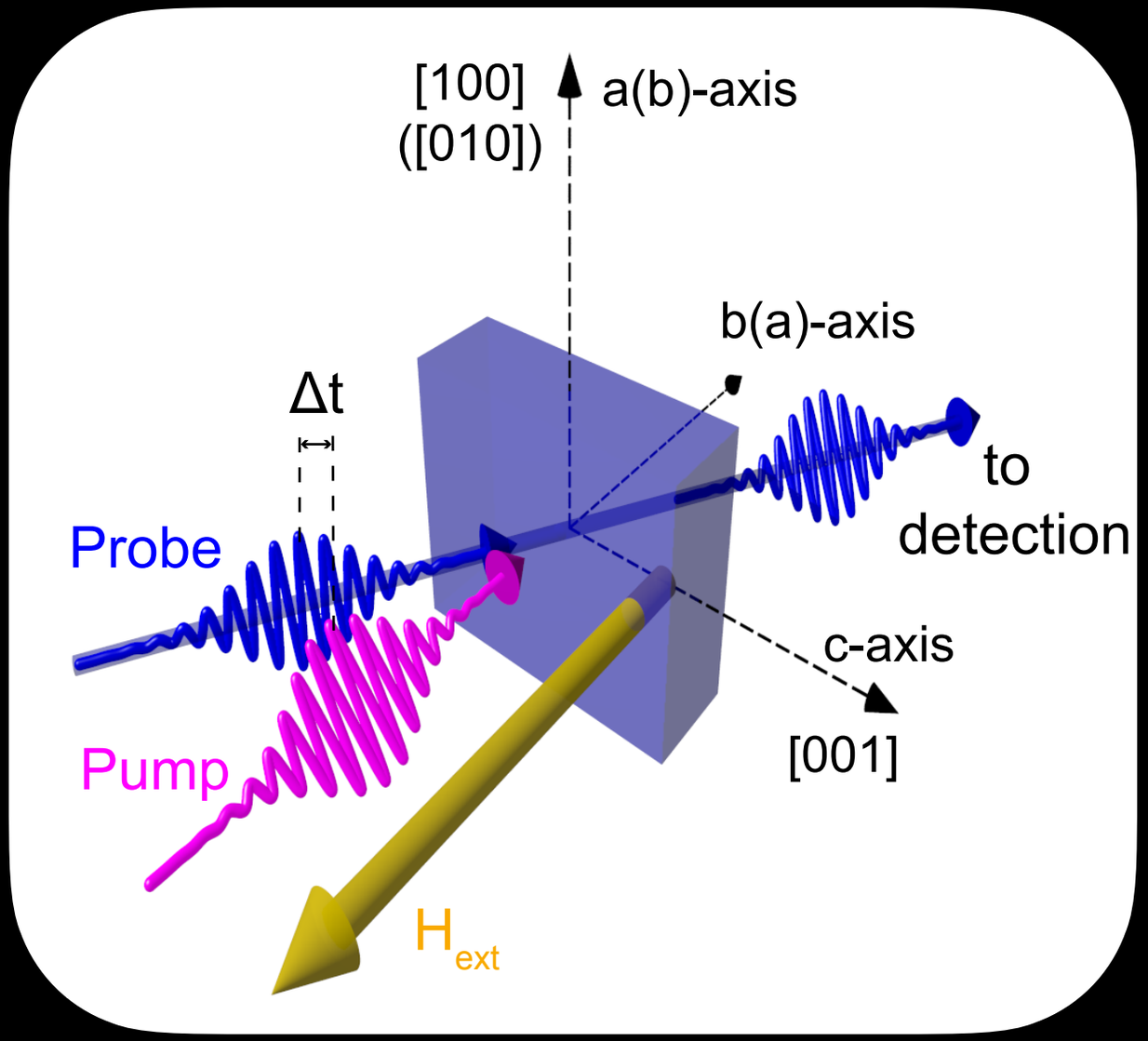
Non-reciprocal directional dichroism (NDD)

Optical ME detection



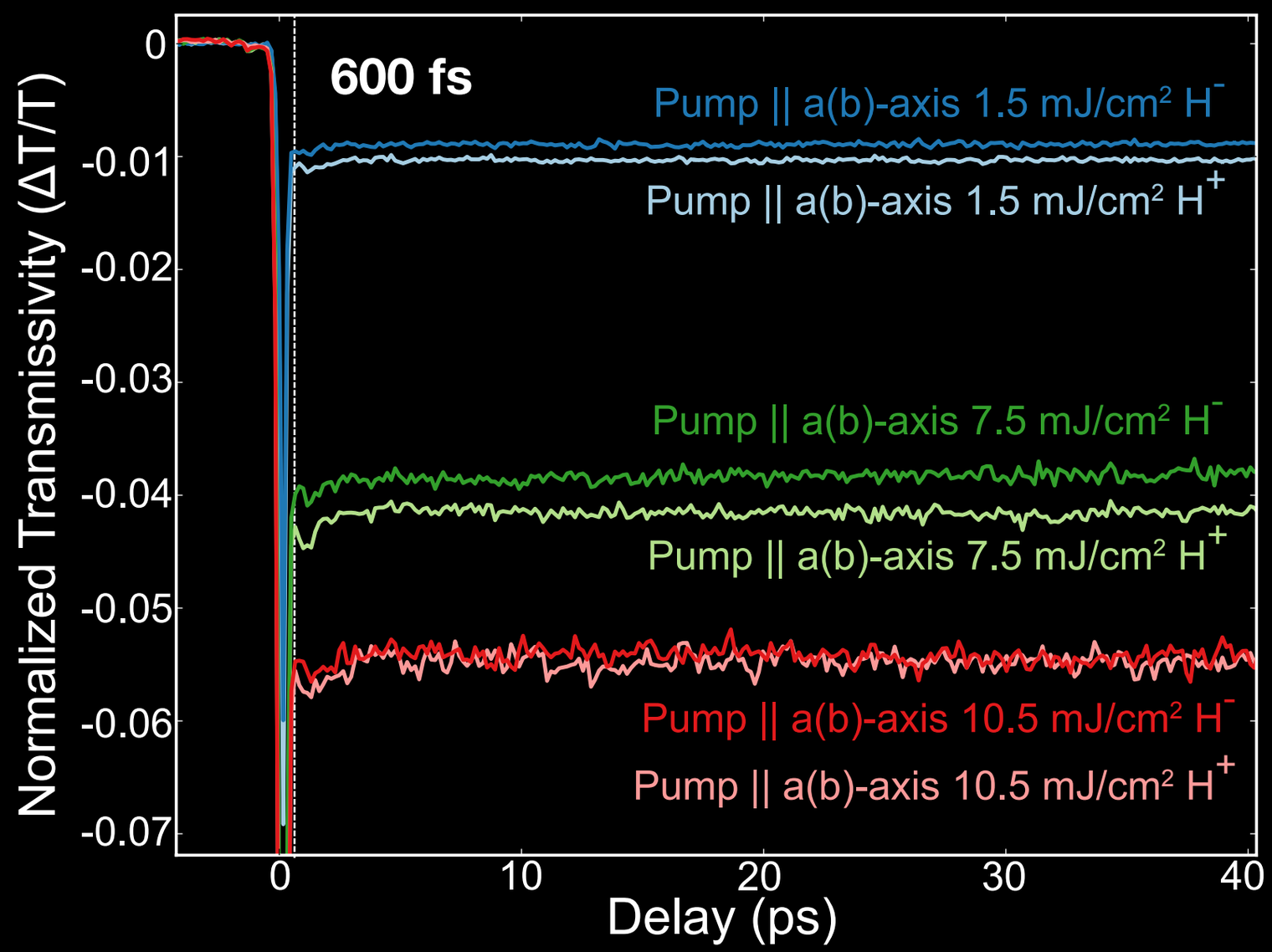
S. Toyoda et al., Phys. Rev. Lett 115, 267207 (2015)

Non-reciprocal directional dichroism (NDD)



Detection: ΔT/T

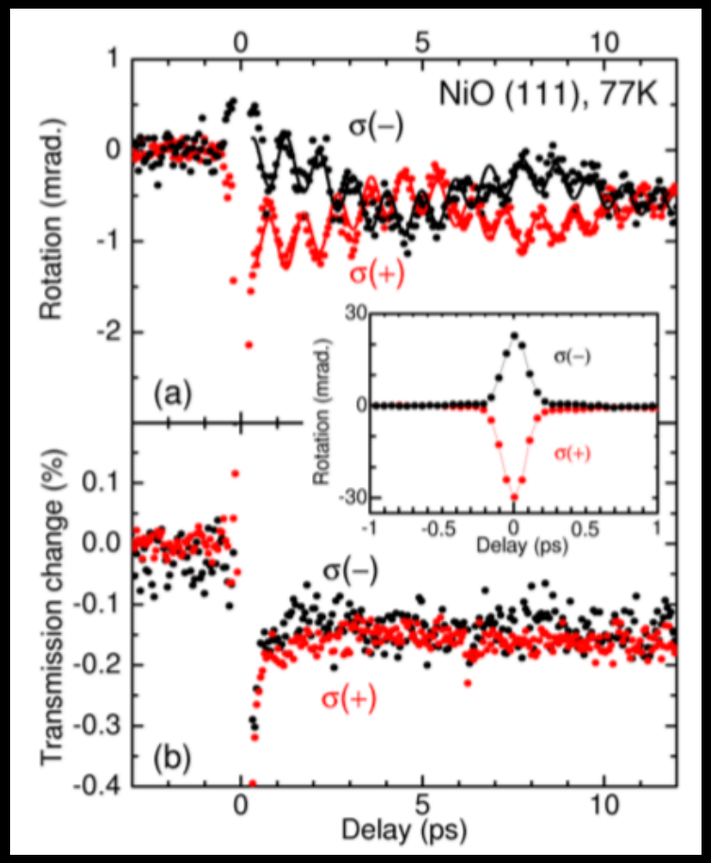
Photo-induce PT



D. Bossini et al. Nature Phys. **14**, 370 (2018)

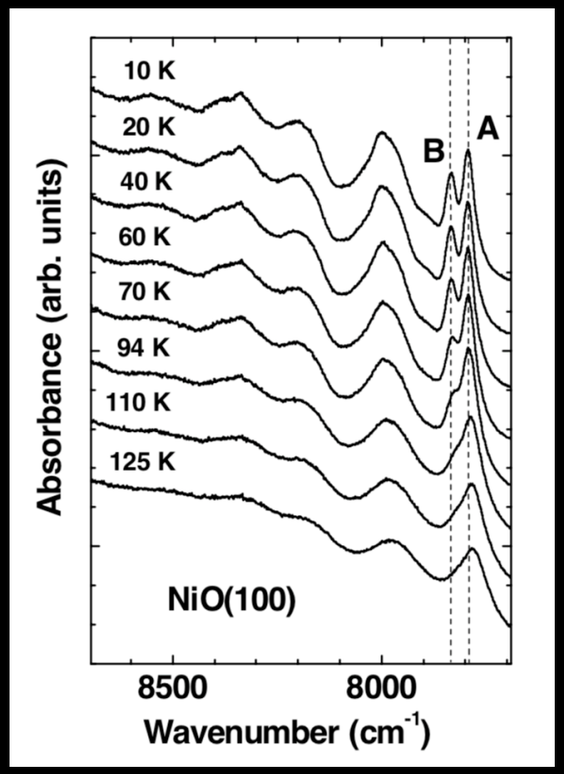
Non-degenerate modes

Non-degenerate modes

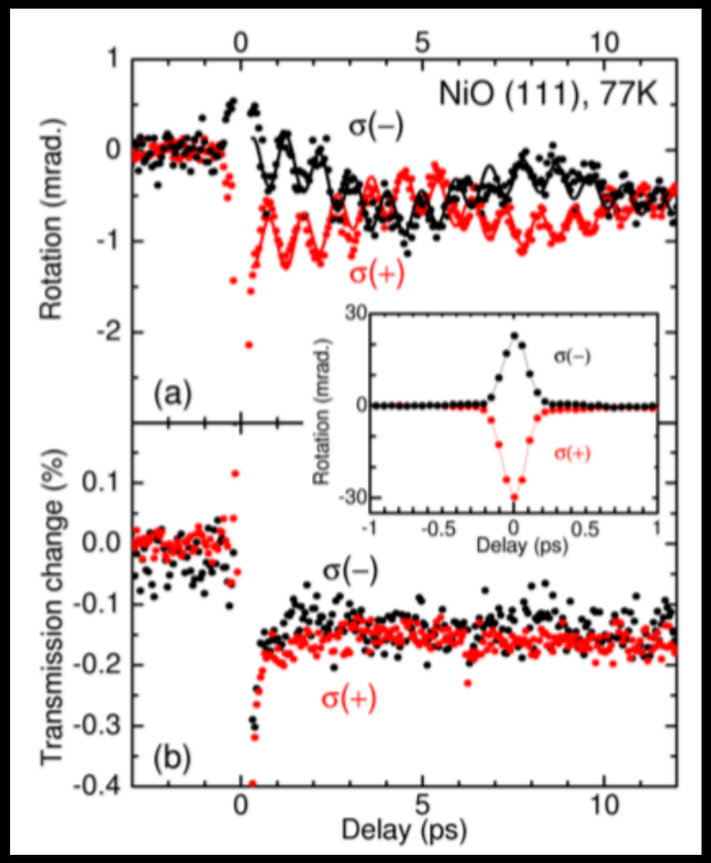


T. Satoh et al PRL **105**, 077402 (2010)

C. Tzschaschel et al PRB **95**, 174407 (2017)



Non-degenerate modes

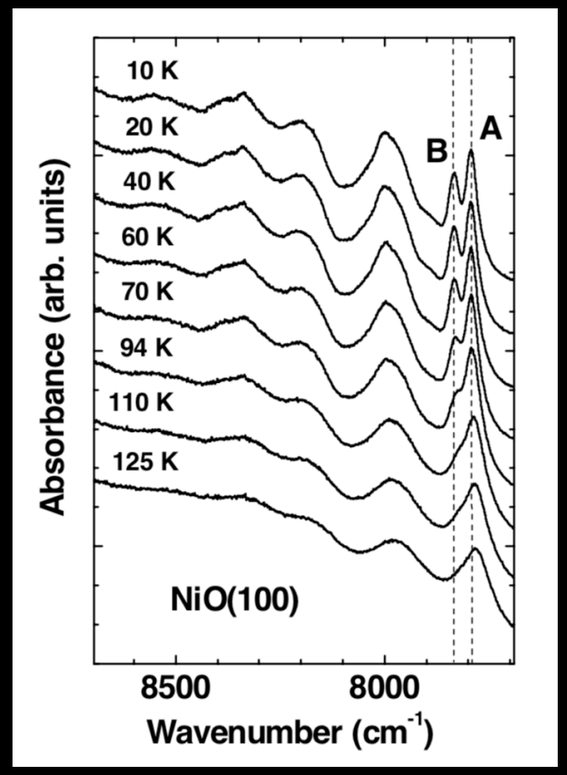


T. Satoh et al PRL **105**, 077402 (2010)

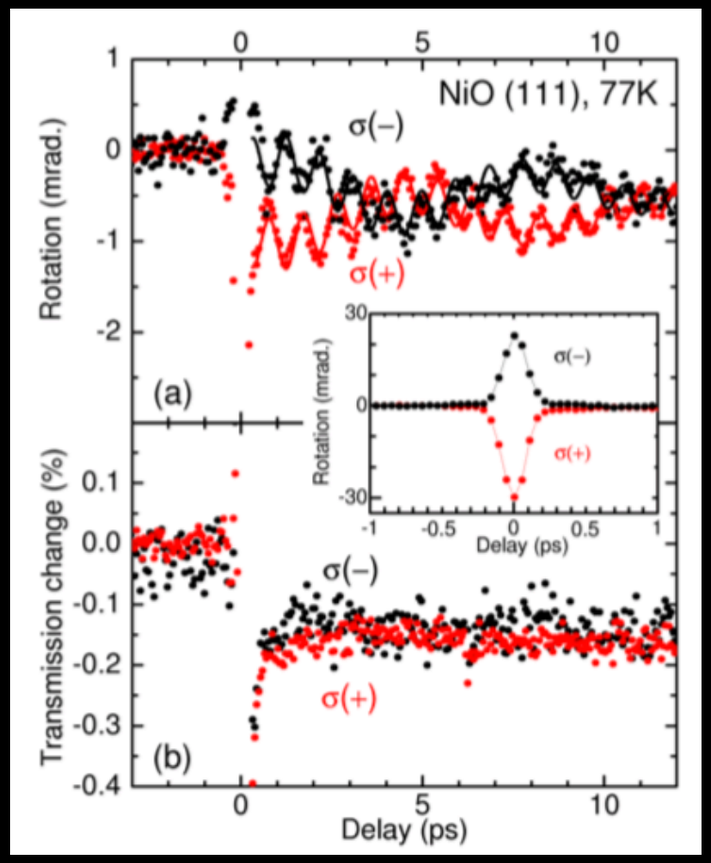
C. Tzschaschel et al PRB **95**, 174407 (2017)

2 modes induced:
110 GHz, 1 THz

X-M: 1 THz mode



Non-degenerate modes



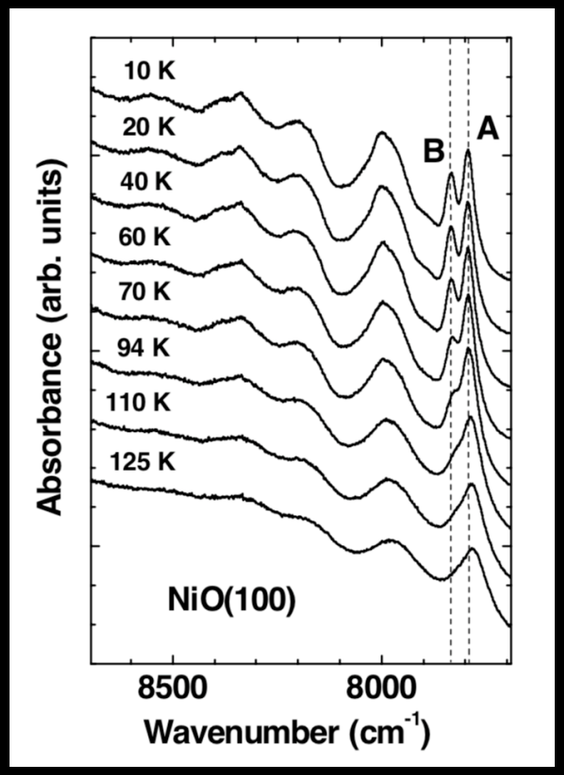
T. Satoh et al PRL **105**, 077402 (2010)

C. Tzschaschel et al PRB **95**, 174407 (2017)

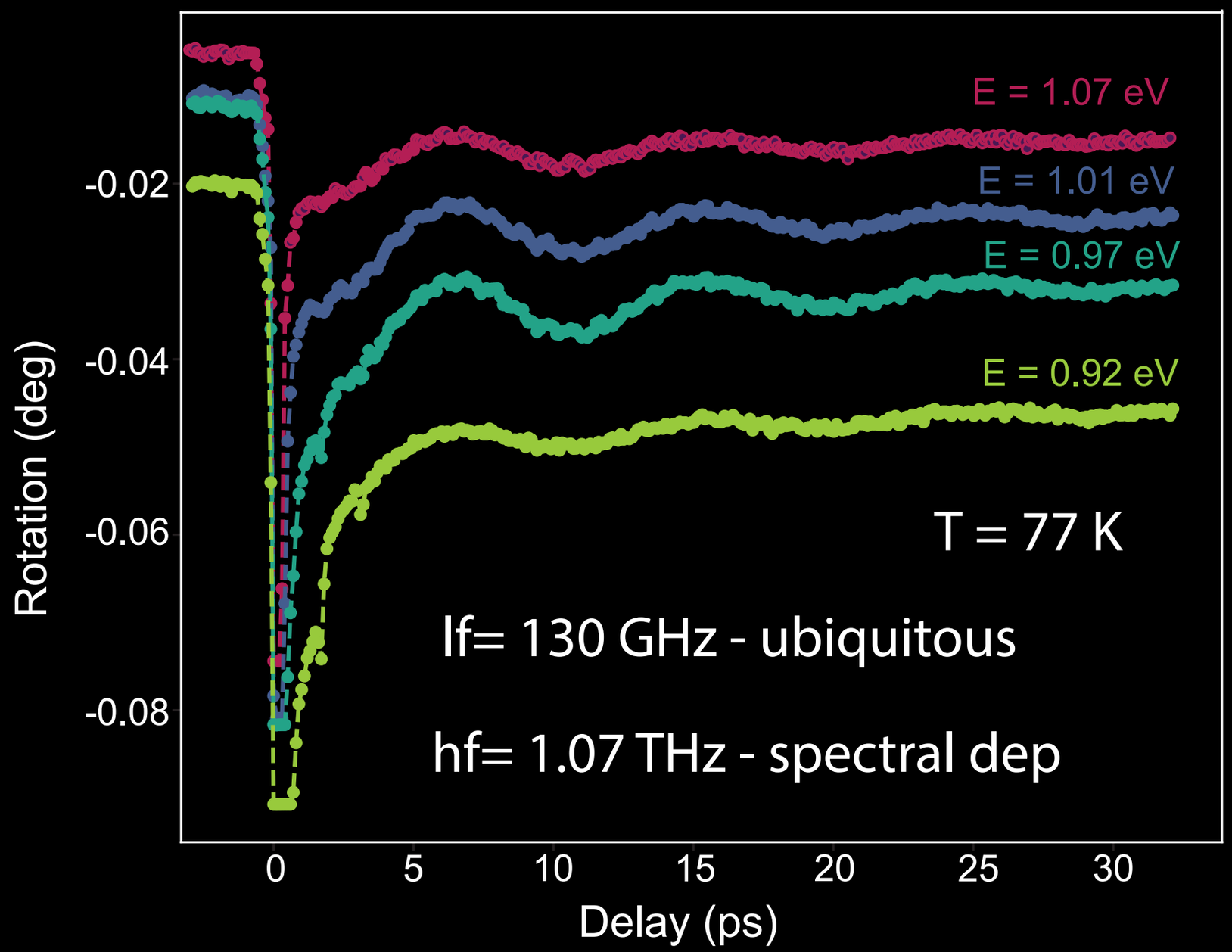
2 modes induced:
110 GHz, 1 THz

Selective mode amplification ?

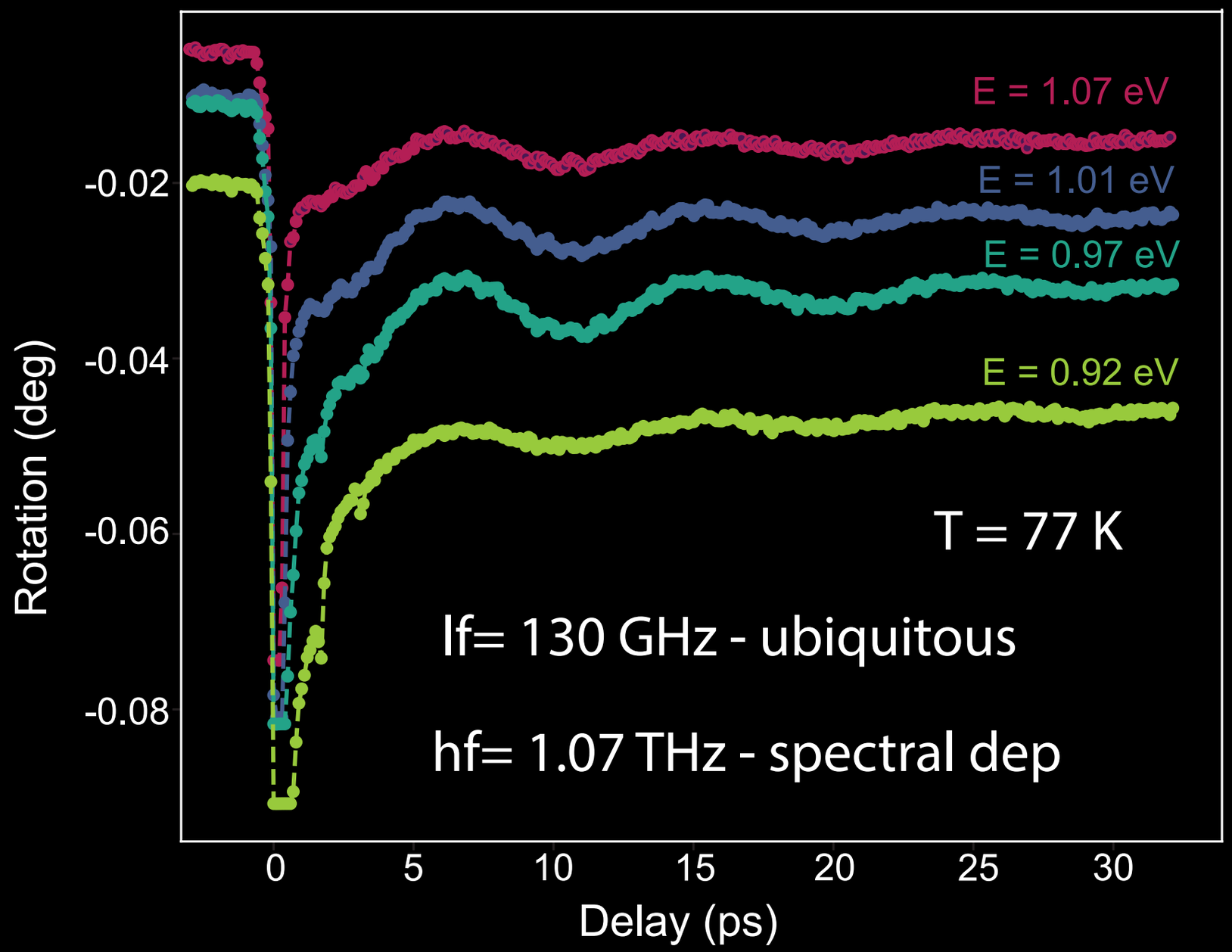
X-M: 1 THz mode

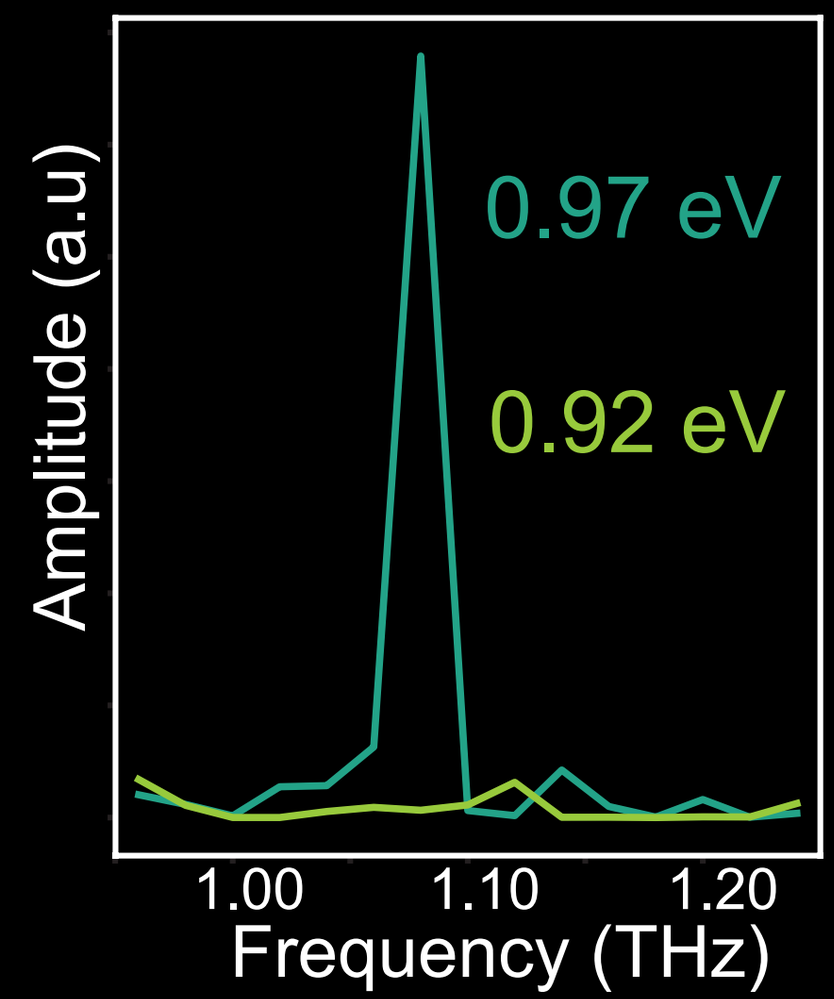


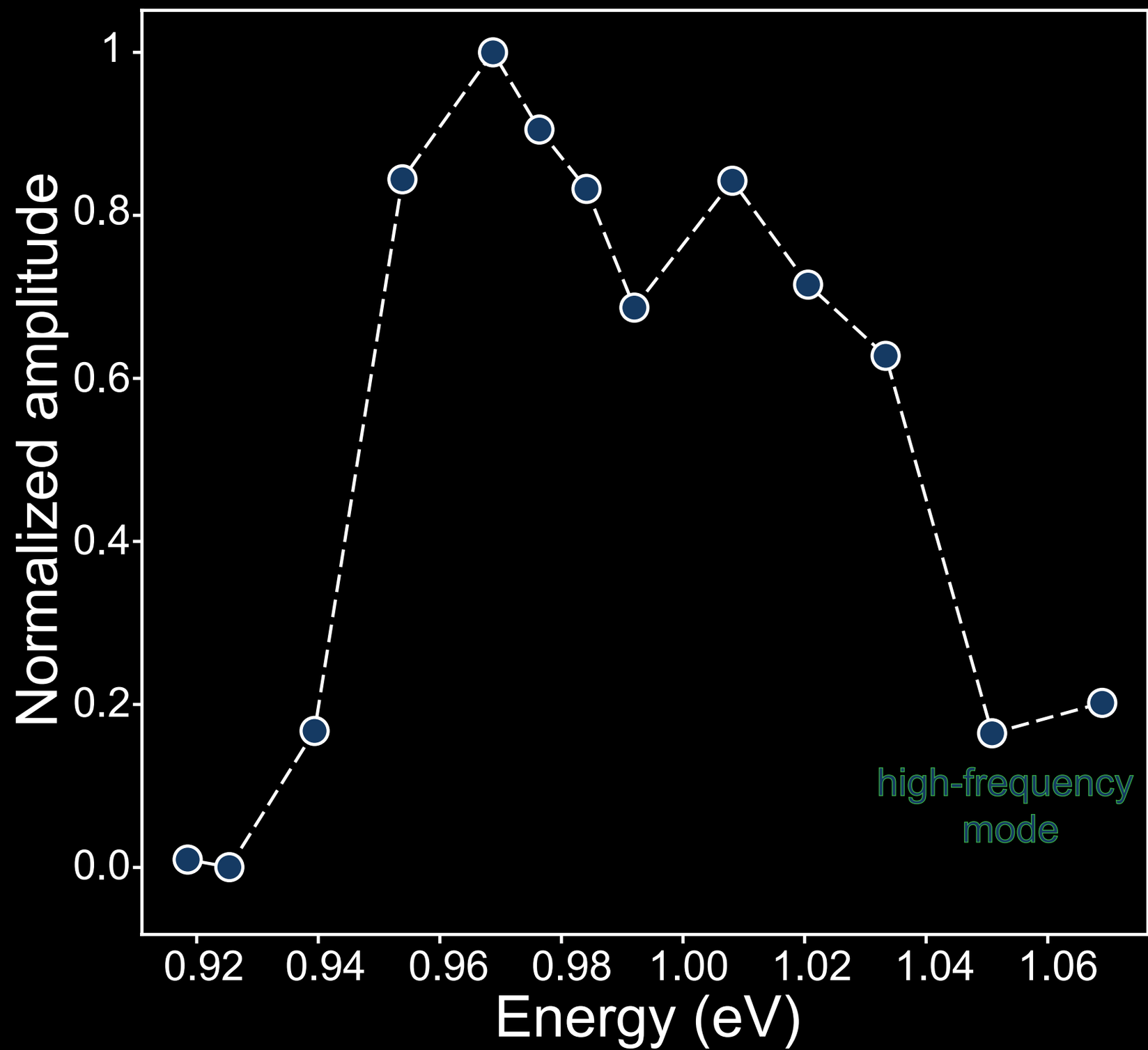
Spin dynamics

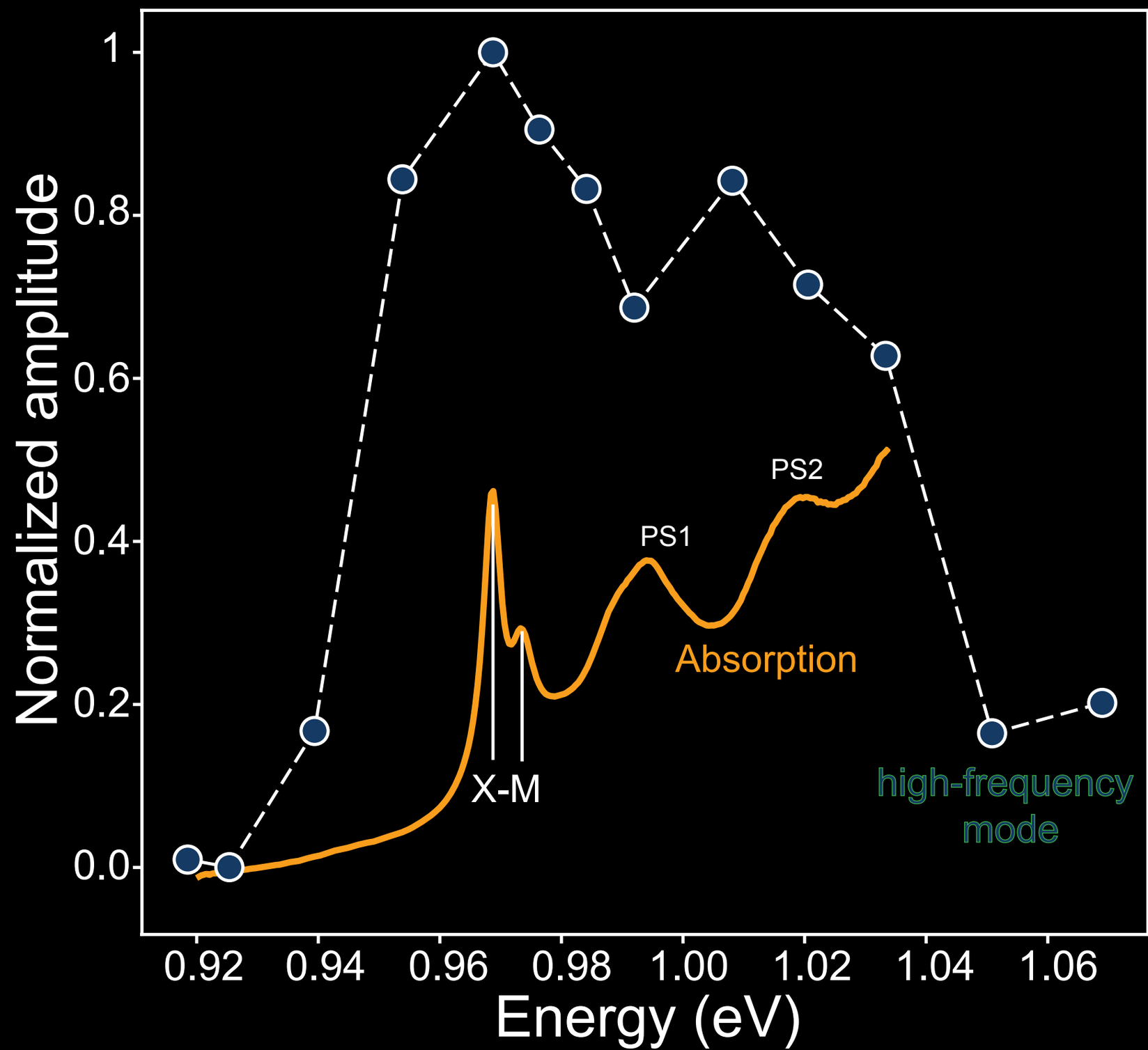


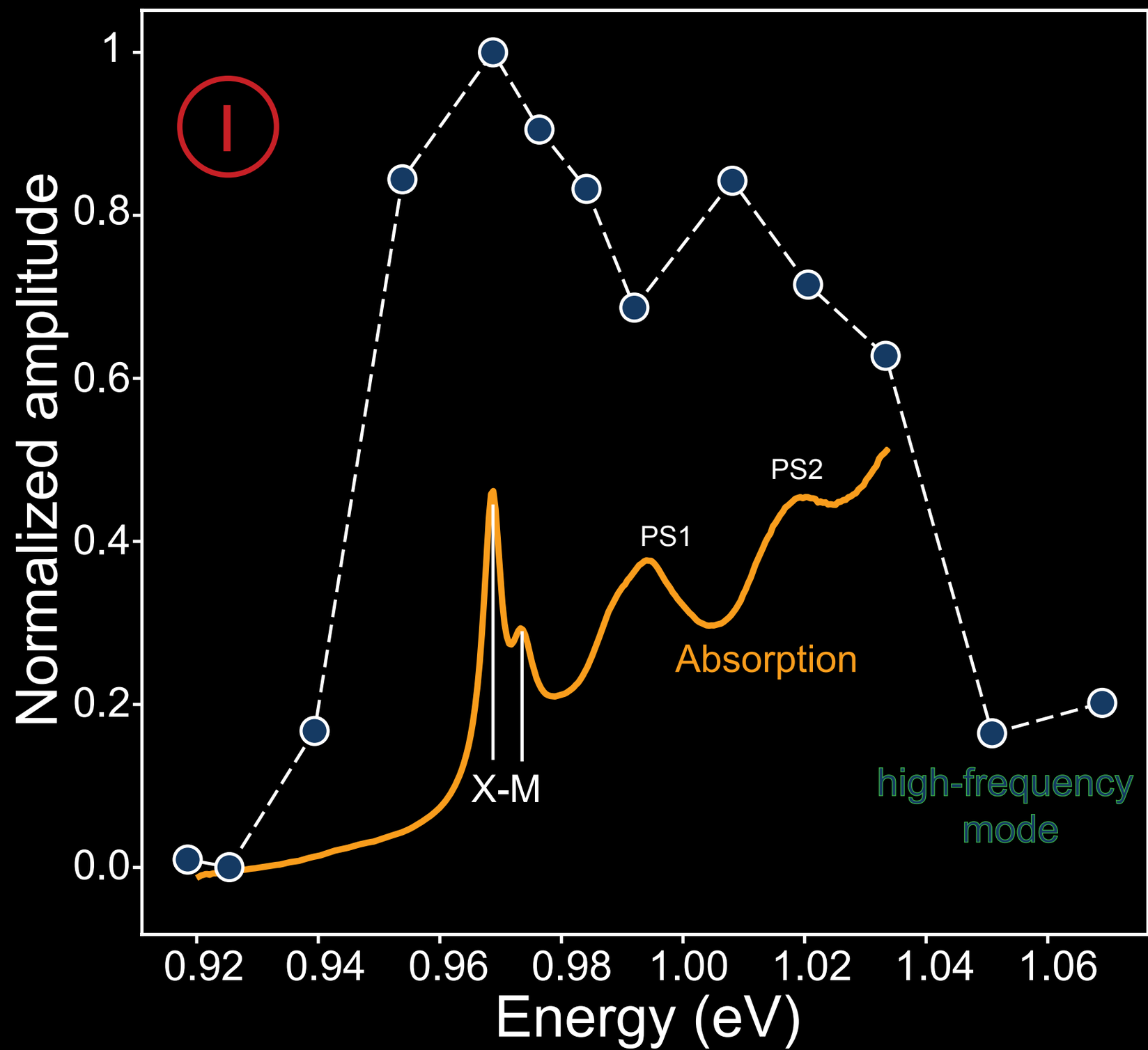
Spin dynamics

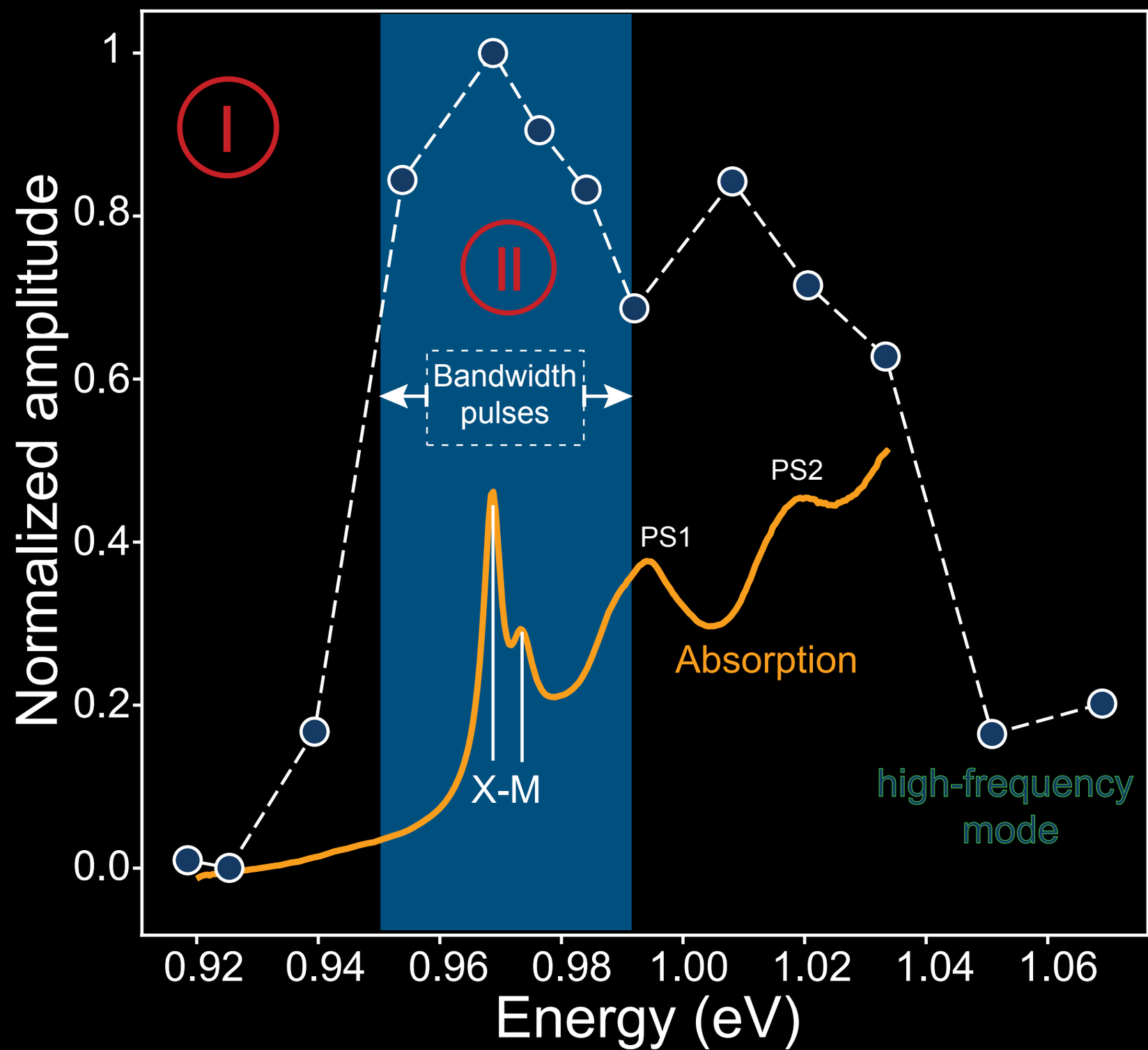


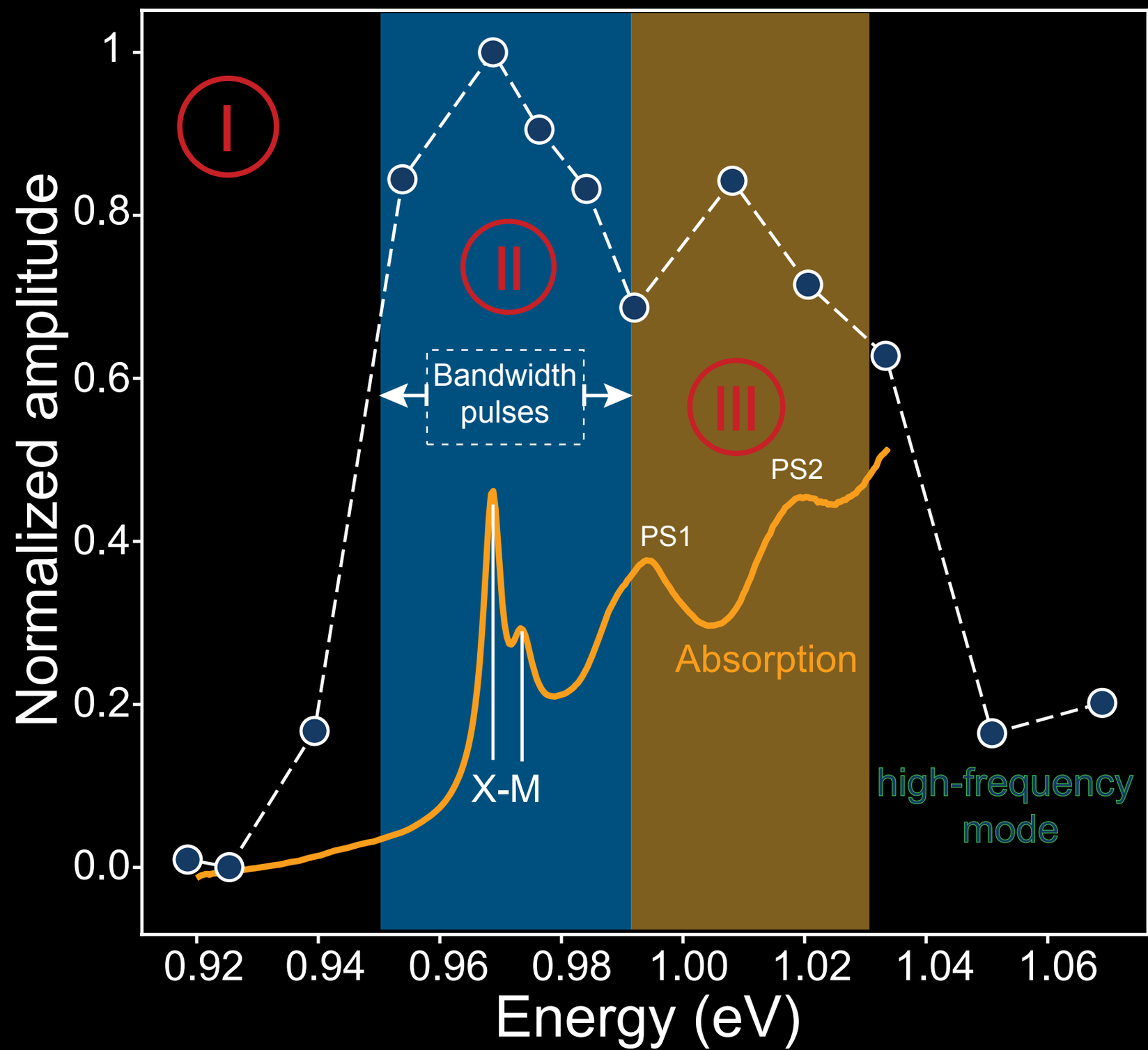


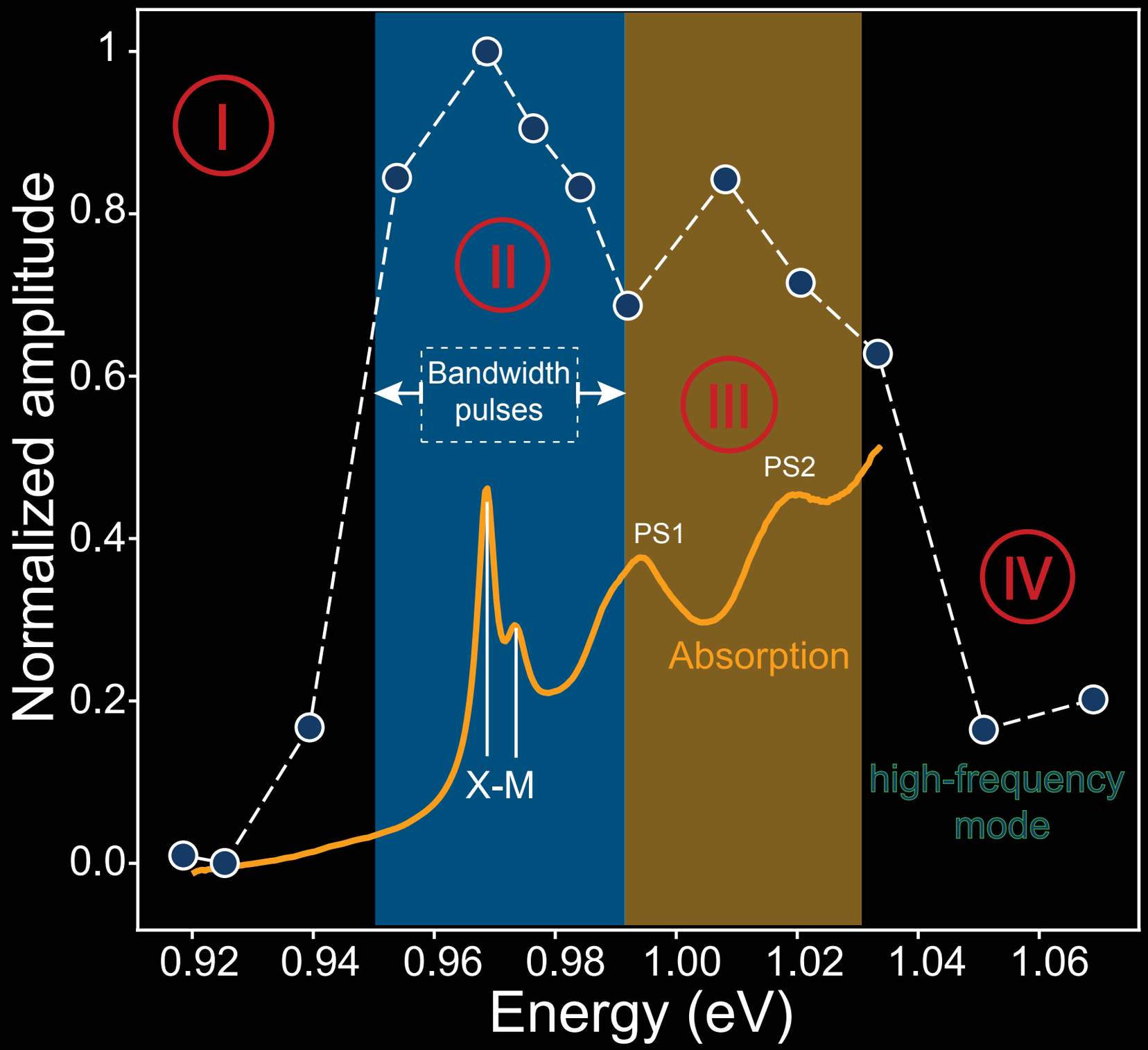

Normalized amplitude
1
0.8
0.6
0.4
0.2
0.0
0.92
0.94
0.96
0.98
1.00
1.02
1.04
1.06
Energy (eV)
high-frequency
mode

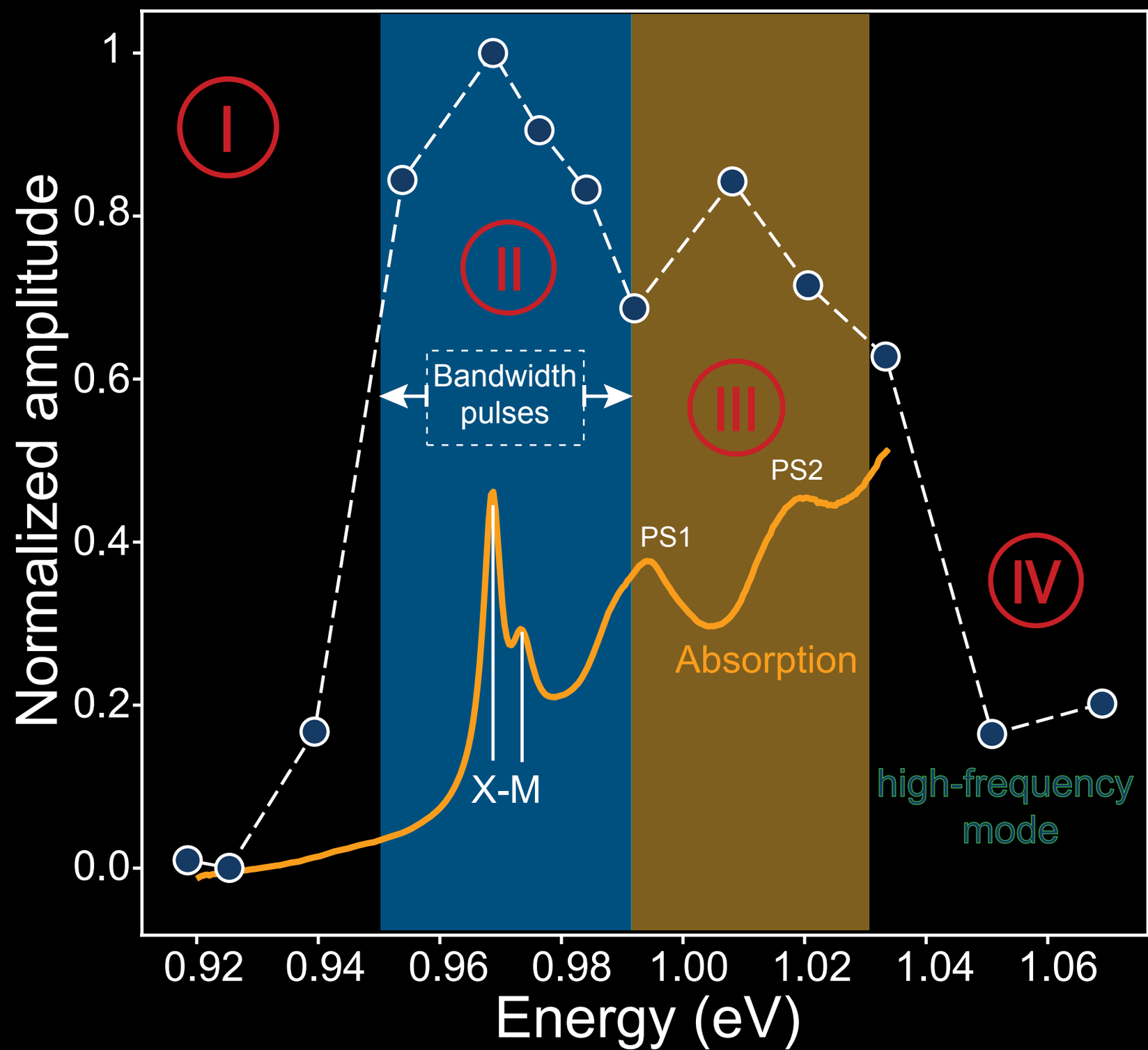

Normalized amplitude
1
0.8
0.6
0.4
0.2
0.0
0.92
0.94
0.96
0.98
1.00
1.02
1.04
1.06
Energy (eV)
PS2
PS1
Absorption
X-M
high-frequency
mode


I
Normalized amplitude
1
0.8
0.6
0.4
0.2
0.0
PS2
PS1
Absorption
X-M
high-frequency
mode
0.92
0.94
0.96
0.98
1.00
1.02
1.04
1.06
Energy (eV)

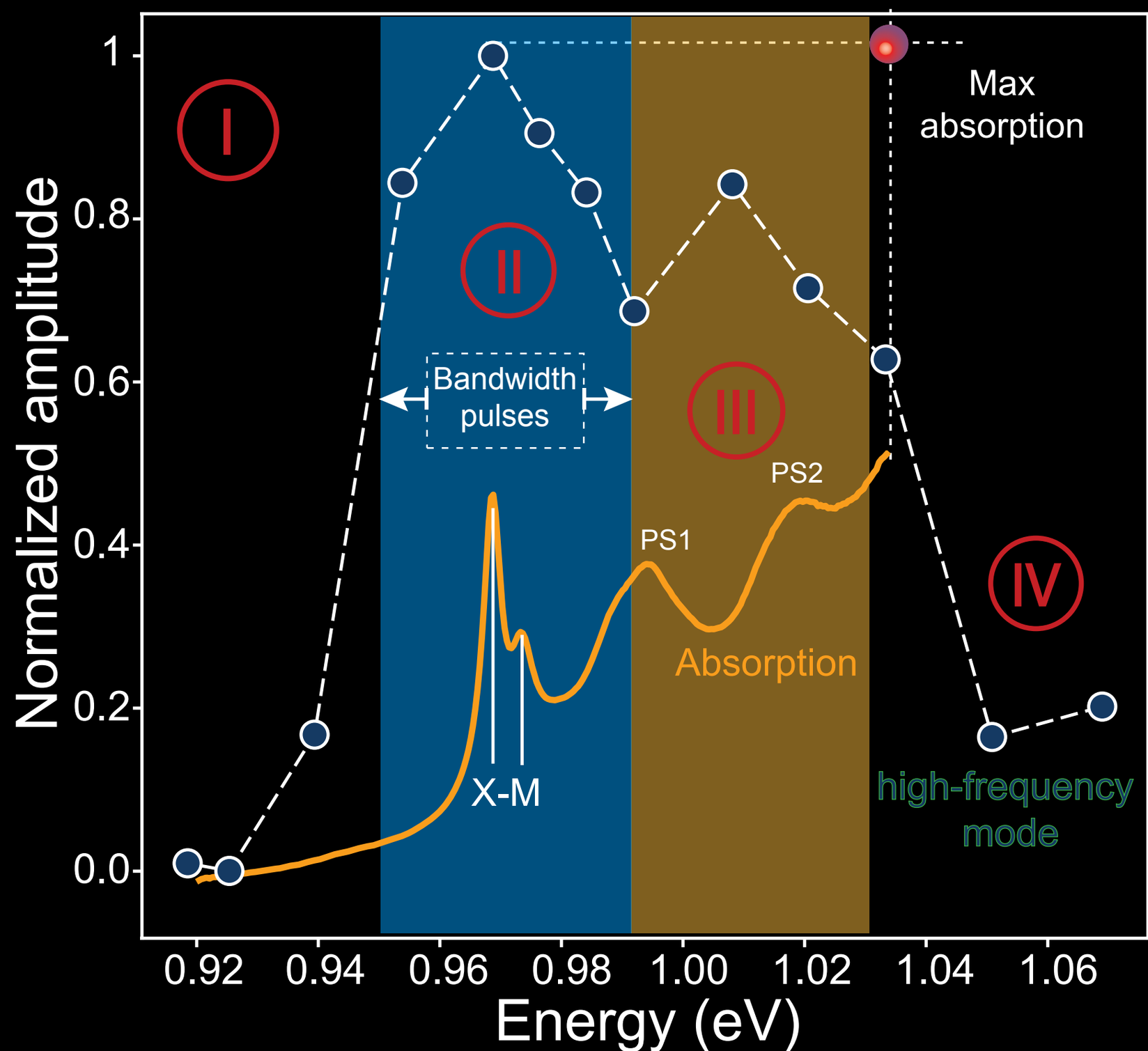

I
II
Bandwidth
pulses
PS2
PS1
Absorption
X-M
high-frequency
mode
Normalized amplitude
1
0.8
0.6
0.4
0.2
0.0
0.92
0.94
0.96
0.98
1.00
1.02
1.04
1.06
Energy (eV)


I
II
III
Bandwidth
pulses
PS2
PS1
Absorption
X-M
high-frequency
mode
Normalized amplitude
Energy (eV)
0.0
0.2
0.4
0.6
0.8
1
0.92
0.94
0.96
0.98
1.00
1.02
1.04
1.06

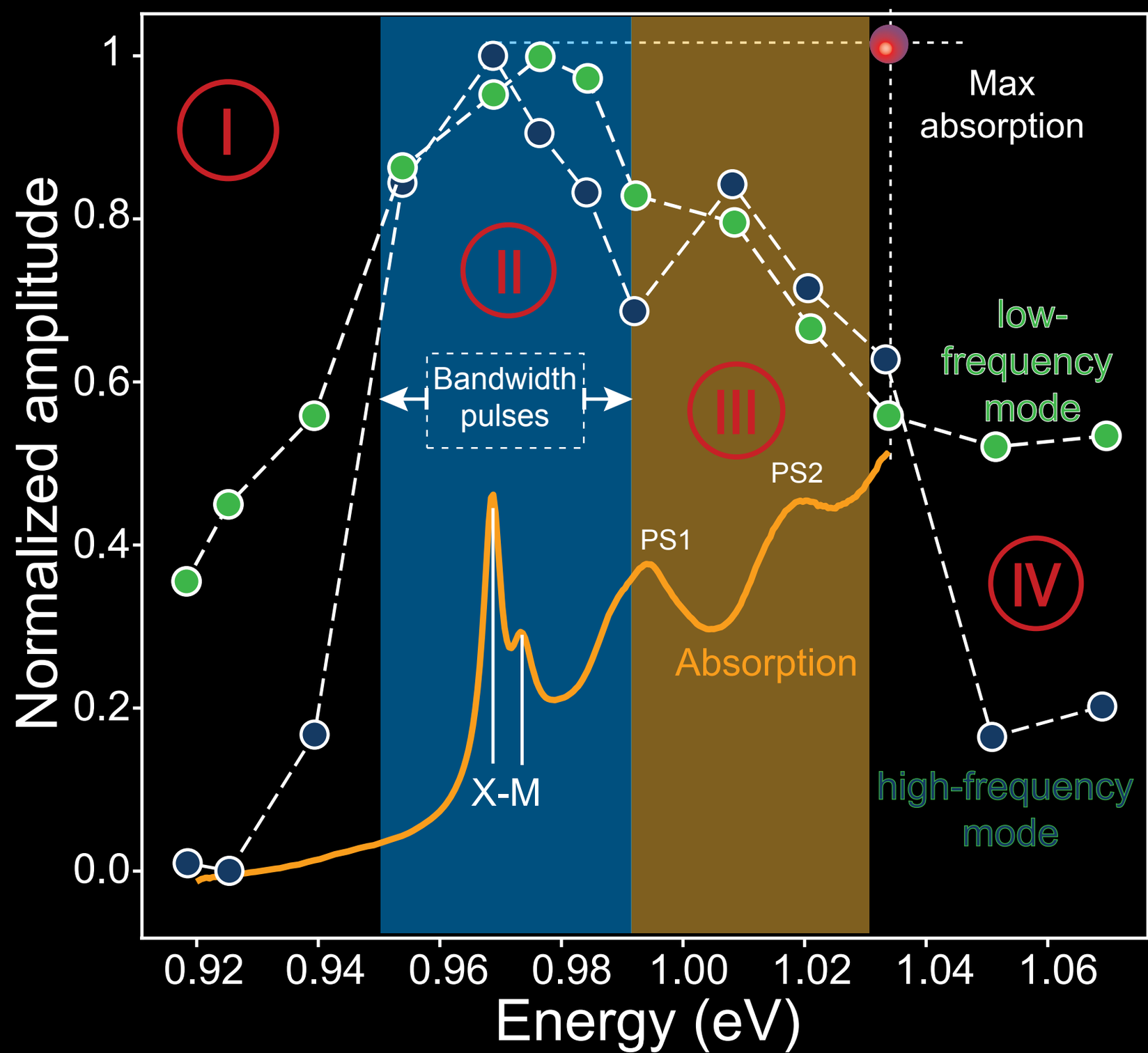

Normalized amplitude
1
0.8
0.6
0.4
0.2
0.0
0.92
0.94
0.96
0.98
1.00
1.02
1.04
1.06
Energy (eV)
I
II
III
IV
Bandwidth
pulses
PS1
PS2
Absorption
X-M
high-frequency
mode



Not purely dissipative mechanism



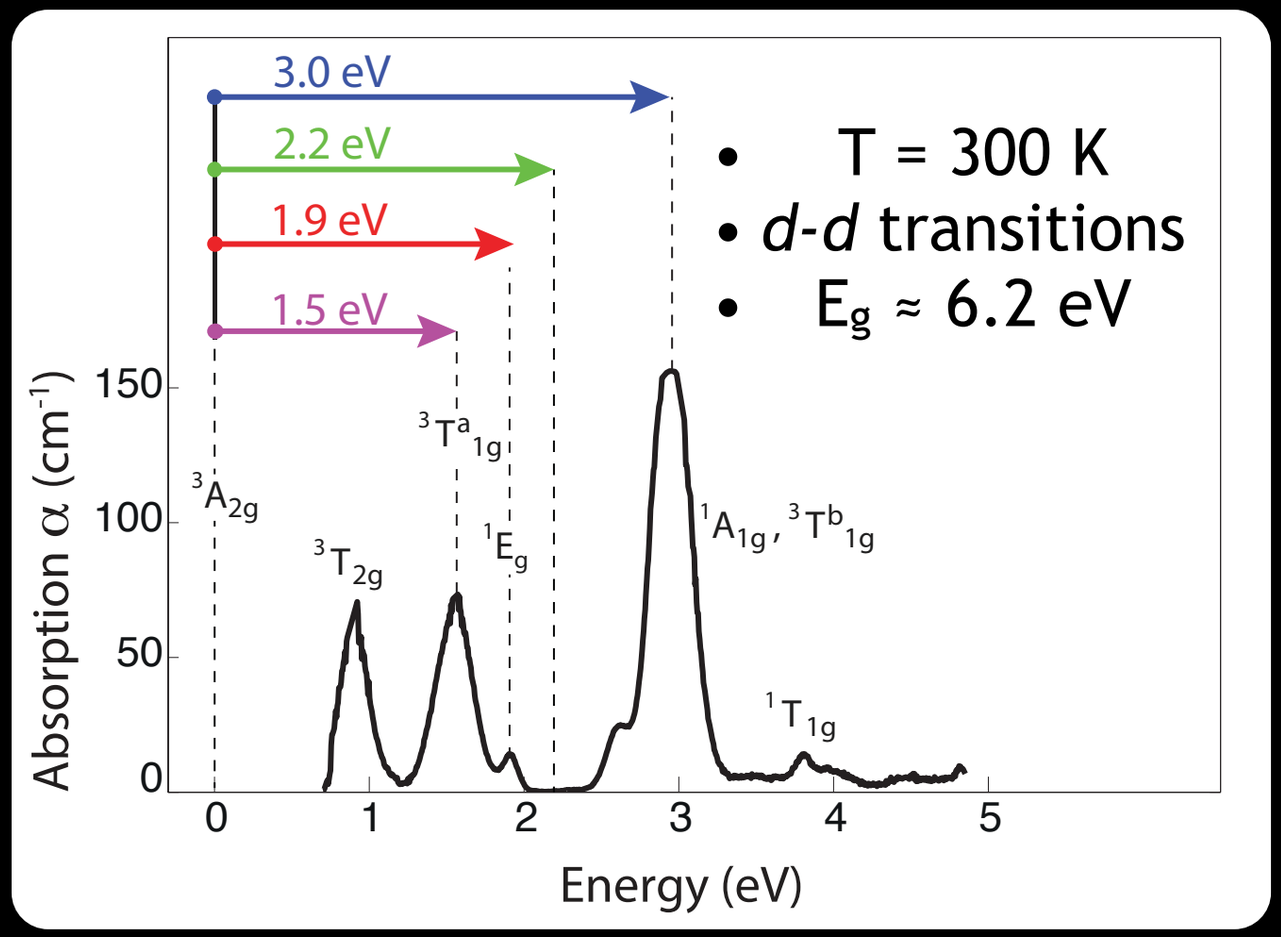
Not purely dissipative mechanism

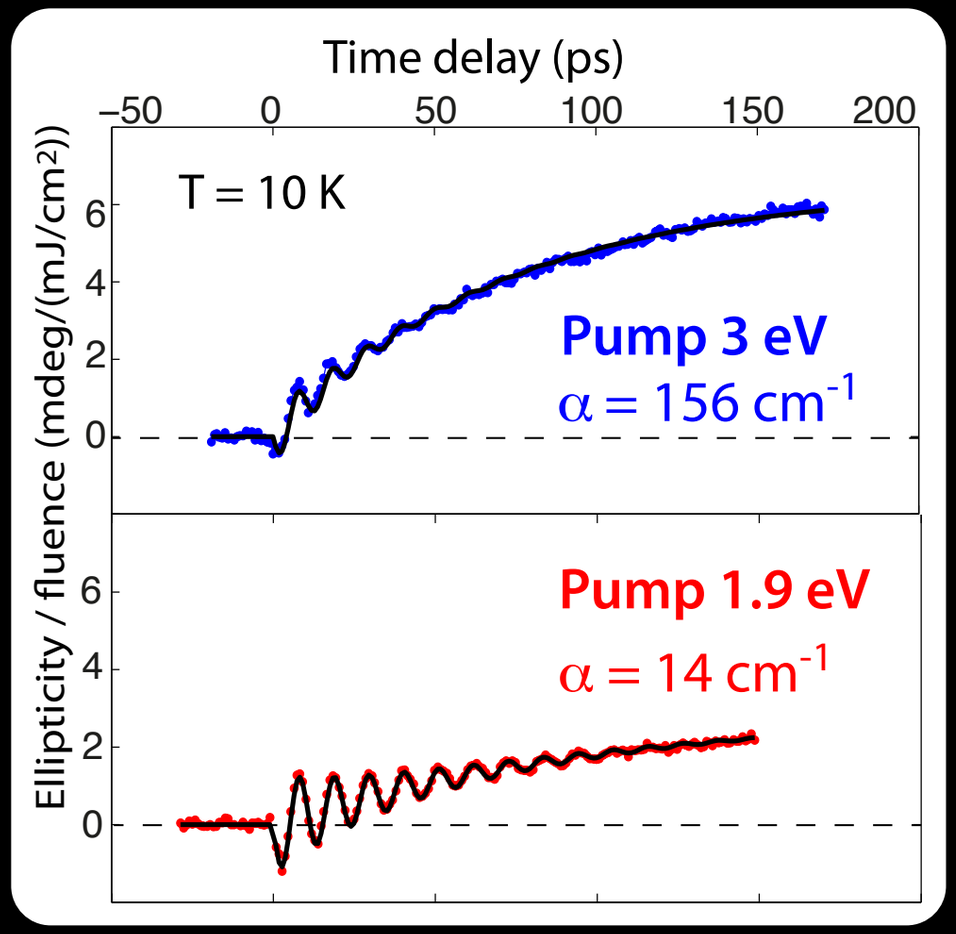


Not purely dissipative mechanism

Unexpected lf-magnons trend

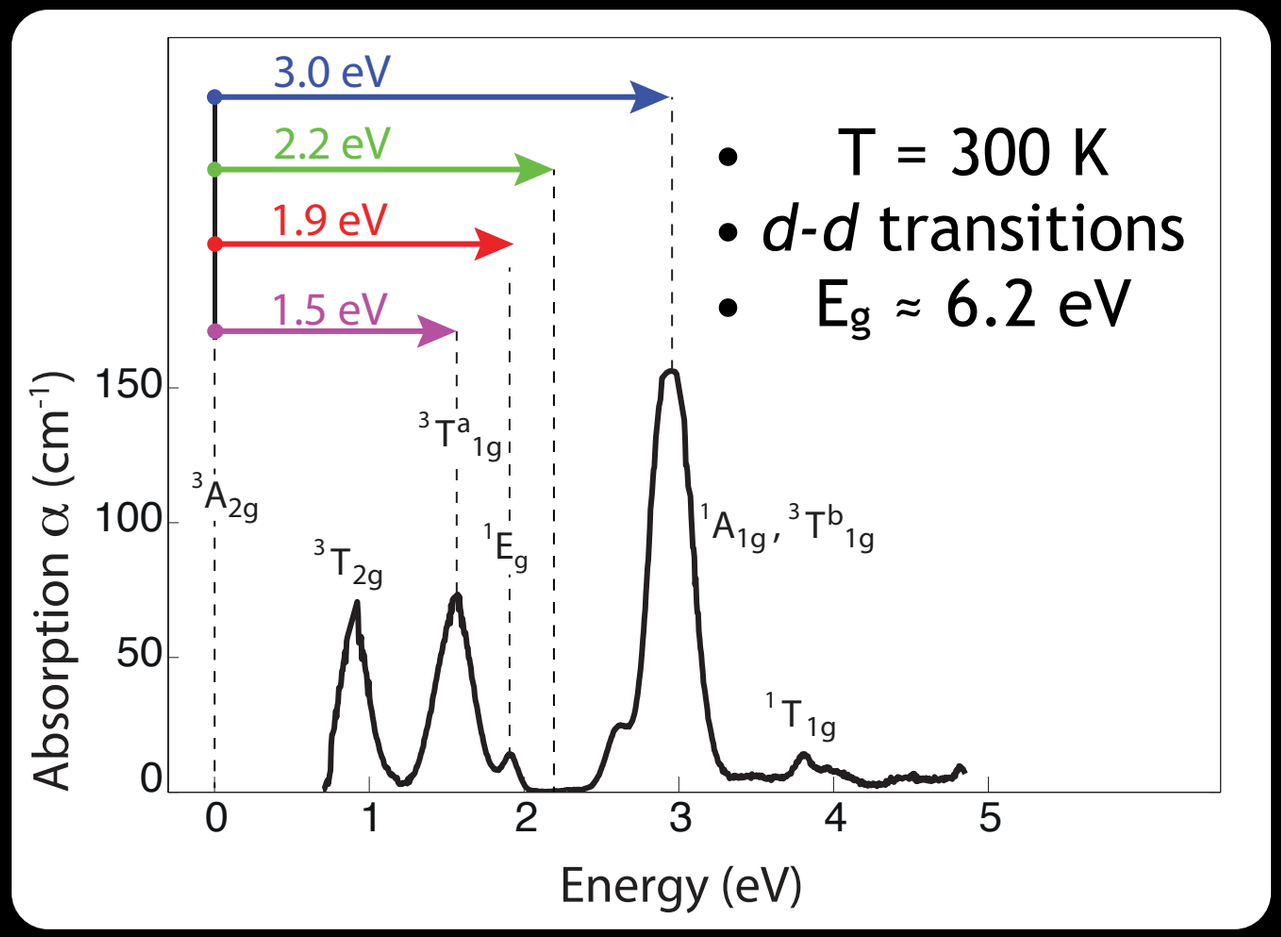
Literature

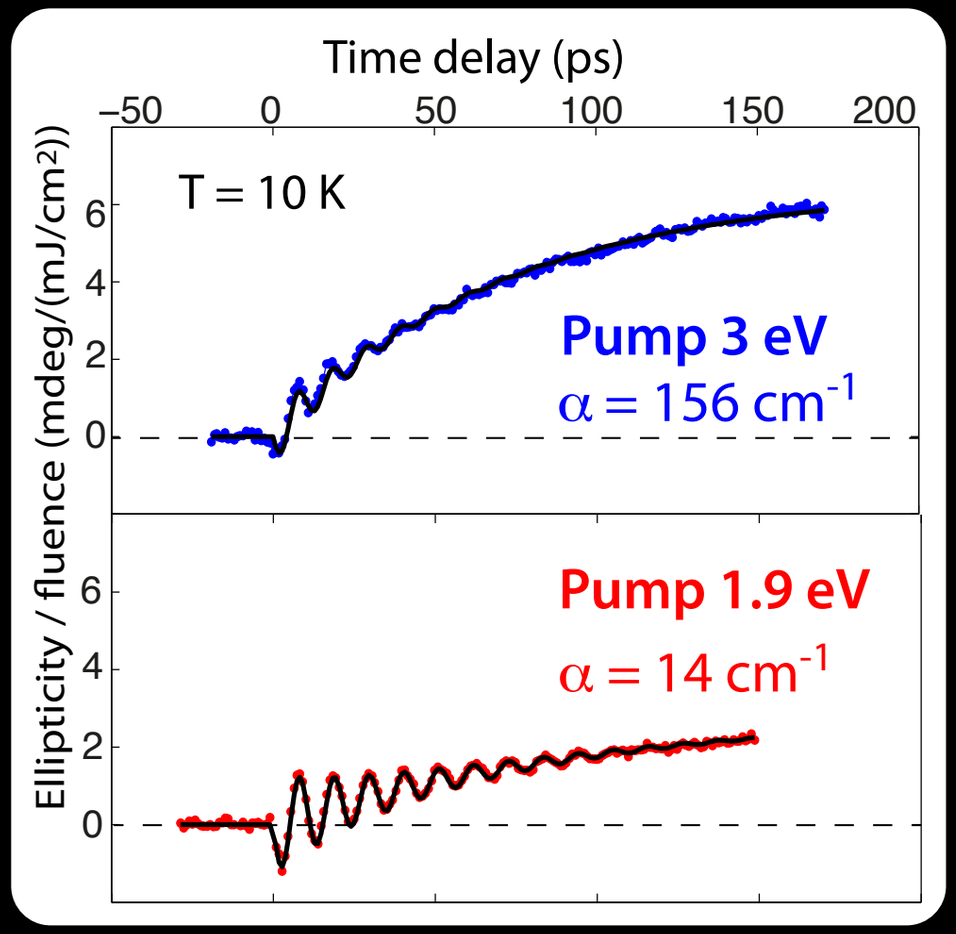




D.Bossini et al PRB. **89** (R), 060405 (2014)

Literature





D.Bossini et al PRB. **89** (R), 060405 (2014)

No spectral dependence amplitude magnons

Interpretation?

Interpretation?

Lf-mode involved in X-M as well

Interpretation?

Lf-mode involved in X-M as well

Hf-mode: A mode
Lf-mode: B mode
C. Tzschaschel et al PRB **95**, 174407

Exciton: A- symmetry
Tsuboi and W Kleemann 1994
J. Phys.: Condens. Matter 6 8625

Interpretation?

Lf-mode involved in X-M as well

Hf-mode: A mode
Lf-mode: B mode
C. Tzschaschel et al PRB **95**, 174407

Exciton: A- symmetry
Tsuboi and W Kleemann 1994
J. Phys.: Condens. Matter 6 8625

Spin-lattice coupling

Interpretation?

Lf-mode involved in X-M as well

Hf-mode: A mode
Lf-mode: B mode
C. Tzschaschel et al PRB **95**, 174407

Exciton: A- symmetry
Tsuboi and W Kleemann 1994
J. Phys.: Condens. Matter 6 8625

Spin-lattice coupling

No proper lattice modes

Reichardt, et al. *Journal of Physics C: Solid State Physics* **8**, 3955–3962 (1975).

Domain walls

✓ Our NiO sample: multi-domain state

✓ S-domains: negligible

C. Tzschaschel et al PRB **95**, 174407

✓ T-Domains

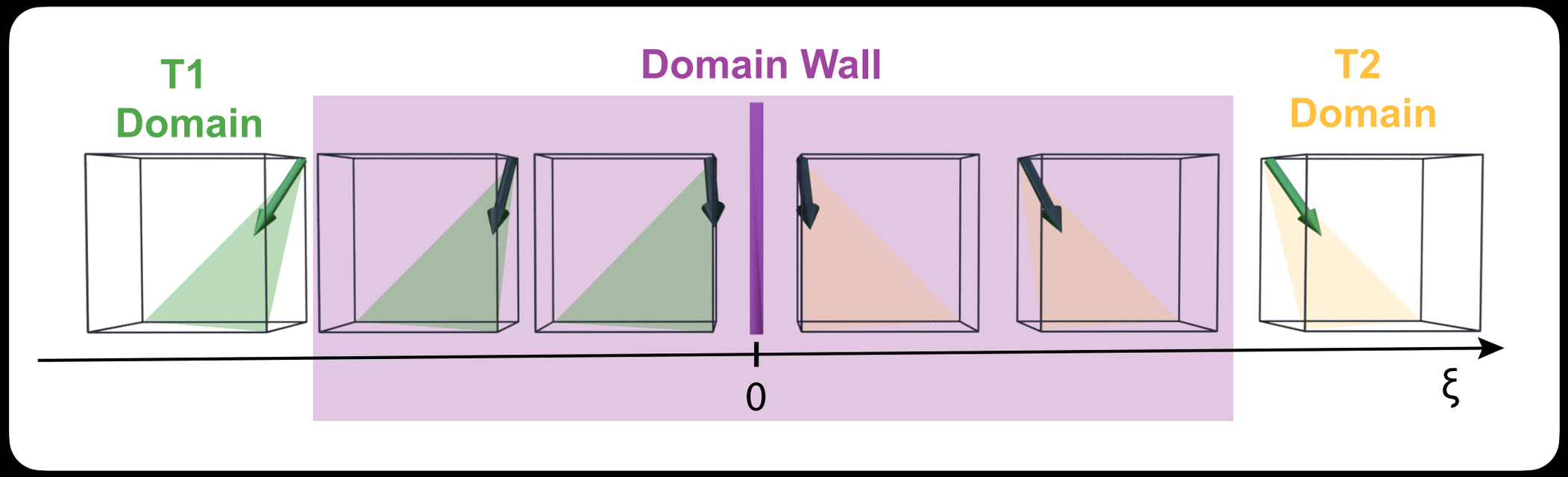
Domain walls

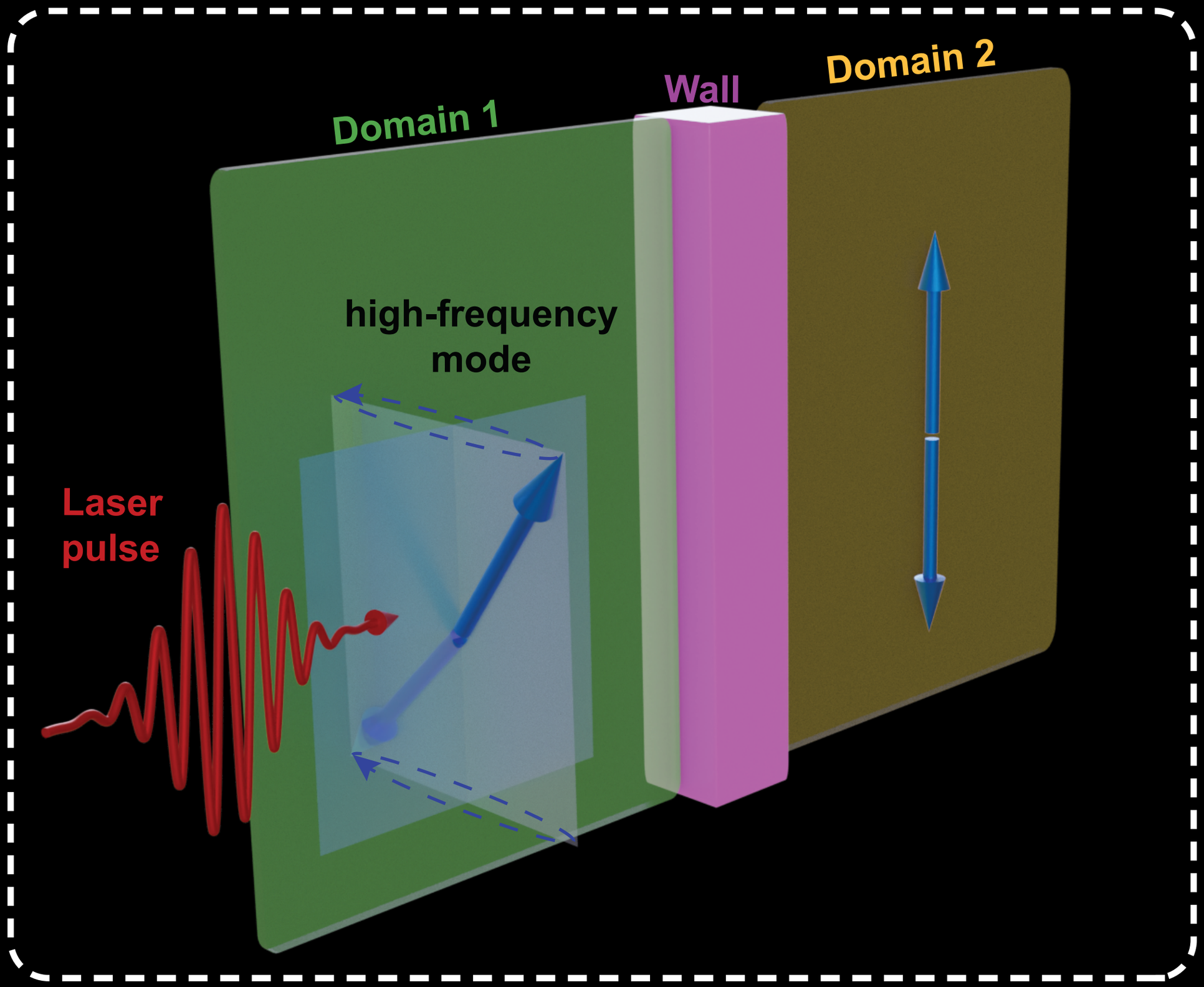
✓ Our NiO sample: multi-domain state

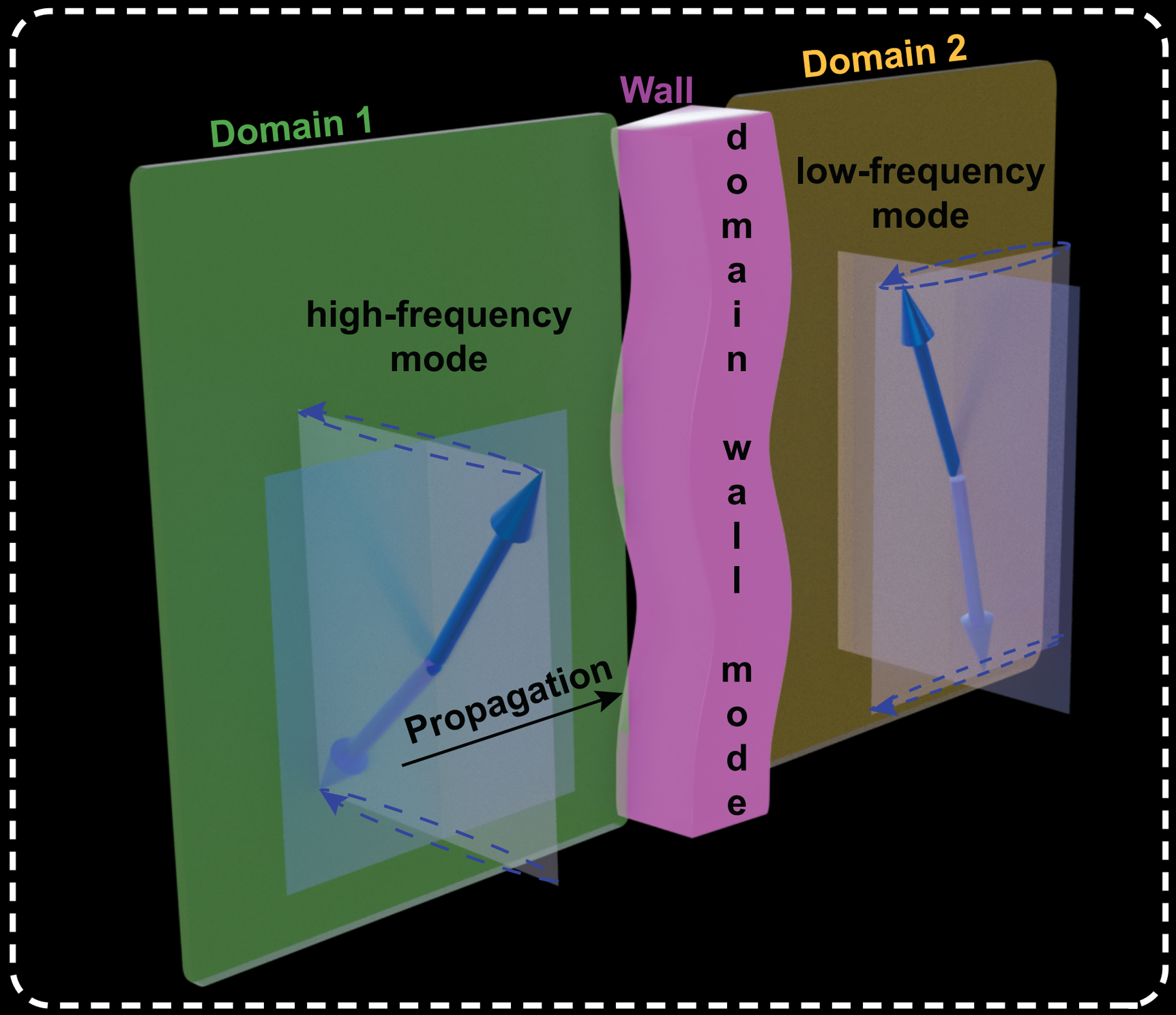
✓ S-domains: negligible

C. Tzschaschel et al PRB **95**, 174407

✓ T-Domains




Domain 1
Wall
Domain 2
high-frequency
mode
Laser
pulse



Wall dynamics (localised mode)

$$\omega_{\mathrm{DW}} \approx \omega_{\mathrm{hf}} + \omega_{lf}$$

Theory

$$\mathbf{n} = a\mathbf{e}_{\mathrm{hf}}(\mathrm{T1})\exp\left(i\omega_{\mathrm{hf}}t\right)$$

Theory

$$\mathbf{n} = a\mathbf{e}_{\mathrm{hf}}(\mathrm{T1})\exp\left(i\omega_{\mathrm{hf}}t\right)$$

I.V. Baryakhtar and B.A. Ivanov *Solid State Communications* **34**, 545 (1980).

$$\mathbf{n}\times\left(\ddot{\mathbf{n}} - c^2\Delta\mathbf{n} + \gamma^2 H_{\mathrm{ex}}\mathbf{H}_{\mathrm{an}}\right) = \omega_{\mathrm{hf}}^2 a\mathbf{n}\times\mathbf{e}_{\mathrm{hf}}(\mathrm{T1})\exp\left(i\omega_{\mathrm{hf}}t\right)\delta(\xi)$$

Theory

$$\mathbf{n} = a\mathbf{e}_{\mathrm{hf}}(\mathrm{T1})\exp\left(i\omega_{\mathrm{hf}}t\right)$$

I.V. Baryakhtar and B.A. Ivanov *Solid State Communications* **34**, 545 (1980).

$$\mathbf{n}\times\left(\ddot{\mathbf{n}} - c^2\Delta\mathbf{n} + \gamma^2 H_{\mathrm{ex}}\mathbf{H}_{\mathrm{an}}\right) = \omega_{\mathrm{hf}}^2 a\mathbf{n}\times\mathbf{e}_{\mathrm{hf}}(\mathrm{T1})\exp\left(i\omega_{\mathrm{hf}}t\right)\delta(\xi)$$

$$\ddot{b}_{\mathrm{DW}} + \frac{1}{\tau}\dot{b}_{\mathrm{DW}} + \omega_{\mathrm{DW}}^2 b_{\mathrm{DW}} = \omega_{\mathrm{hf}}^2 a\exp\left(i\omega_{\mathrm{hf}}t\right),$$

$$\ddot{b}_{\mathrm{hf}} + \frac{1}{\tau}\dot{b}_{\mathrm{hf}} + \omega_{\mathrm{hf}}^2 b_{\mathrm{hf}} = \frac{1}{3}\omega_{\mathrm{hf}}^2 a\exp\left(i\omega_{\mathrm{hf}}t\right),$$

$$\ddot{b}_{\mathrm{lf}} + \frac{1}{\tau}\dot{b}_{\mathrm{lf}} + \omega_{\mathrm{lf}}^2\left(1 - 4b_{\mathrm{DW}}\right)b_{\mathrm{lf}} = \frac{1}{3}\omega_{\mathrm{hf}}^2 a\exp\left(i\omega_{\mathrm{hf}}t\right)$$

Theory

$$\mathbf{n} = a\mathbf{e}_{\mathrm{hf}}(\mathrm{T1})\exp\left(i\omega_{\mathrm{hf}}t\right)$$

I.V. Baryakhtar and B.A. Ivanov *Solid State Communications* **34**, 545 (1980).

$$\mathbf{n}\times\left(\ddot{\mathbf{n}} - c^2\Delta\mathbf{n} + \gamma^2 H_{\mathrm{ex}}\mathbf{H}_{\mathrm{an}}\right) = \omega_{\mathrm{hf}}^2 a\mathbf{n}\times\mathbf{e}_{\mathrm{hf}}(\mathrm{T1})\exp\left(i\omega_{\mathrm{hf}}t\right)\delta(\xi)$$

$$\ddot{b}_{\mathrm{DW}} + \frac{1}{\tau}\dot{b}_{\mathrm{DW}} + \omega_{\mathrm{DW}}^2 b_{\mathrm{DW}} = \omega_{\mathrm{hf}}^2 a\exp\left(i\omega_{\mathrm{hf}}t\right),$$

$$\ddot{b}_{\mathrm{hf}} + \frac{1}{\tau}\dot{b}_{\mathrm{hf}} + \omega_{\mathrm{hf}}^2 b_{\mathrm{hf}} = \frac{1}{3}\omega_{\mathrm{hf}}^2 a\exp\left(i\omega_{\mathrm{hf}}t\right),$$

$$\ddot{b}_{\mathrm{lf}} + \frac{1}{\tau}\dot{b}_{\mathrm{lf}} + \omega_{\mathrm{lf}}^2\left(1 - 4b_{\mathrm{DW}}\right)b_{\mathrm{lf}} = \frac{1}{3}\omega_{\mathrm{hf}}^2 a\exp\left(i\omega_{\mathrm{hf}}t\right)$$

Theory

$$\mathbf{n} = a\mathbf{e}_{\mathrm{hf}}(\mathrm{T1})\exp\left(i\omega_{\mathrm{hf}}t\right)$$

I.V. Baryakhtar and B.A. Ivanov *Solid State Communications* **34**, 545 (1980).

$$\mathbf{n}\times\left(\ddot{\mathbf{n}} - c^2\Delta\mathbf{n} + \gamma^2 H_{\mathrm{ex}}\mathbf{H}_{\mathrm{an}}\right) = \omega_{\mathrm{hf}}^2 a\mathbf{n}\times\mathbf{e}_{\mathrm{hf}}(\mathrm{T1})\exp\left(i\omega_{\mathrm{hf}}t\right)\delta(\xi)$$

$$\ddot{b}_{\mathrm{D}}$$

$$b_{\mathrm{lf}} = \frac{a^2 e^{\lambda t}}{3}\exp\left(i\omega_{\mathrm{lf}}t\right)$$

$$t),$$

$$_{\mathrm{hf}}t),$$

$$\ddot{b}_{\mathrm{lf}} + \frac{1}{\tau}\dot{b}_{\mathrm{lf}} + \omega_{\mathrm{lf}}^2\left(1 - 4b_{\mathrm{DW}}\right)b_{\mathrm{lf}} = \frac{1}{3}\omega_{\mathrm{hf}}^2 a\exp\left(i\omega_{\mathrm{hf}}t\right)$$

Theory

$$\mathbf{n} = a\mathbf{e}_{\mathrm{hf}}(\mathrm{T1})\exp\left(i\omega_{\mathrm{hf}}t\right)$$

I.V. Baryakhtar and B.A. Ivanov *Solid State Communications* **34**, 545 (1980).

$$\mathbf{n}\times\left(\ddot{\mathbf{n}} - c^2\Delta\mathbf{n} + \gamma^2 H_{\mathrm{ex}}\mathbf{H}_{\mathrm{an}}\right) = \omega_{\mathrm{hf}}^2 a\mathbf{n}\times\mathbf{e}_{\mathrm{hf}}(\mathrm{T1})\exp\left(i\omega_{\mathrm{hf}}t\right)\delta(\xi)$$

$$\ddot{b}_{\mathrm{D}}$$

$$b_{\mathrm{lf}} = \frac{a^2 e^{\lambda t}}{3}\exp\left(i\omega_{\mathrm{lf}}t\right)$$

$$t),$$

$$_{\mathrm{hf}}t),$$

$$\ddot{b}_{\mathrm{lf}} + \frac{1}{\tau}\dot{b}_{\mathrm{lf}} + \omega_{\mathrm{lf}}^2\left(1 - 4b_{\mathrm{DW}}\right)b_{\mathrm{lf}} = \frac{1}{3}\omega_{\mathrm{hf}}^2 a\exp\left(i\omega_{\mathrm{hf}}t\right)$$

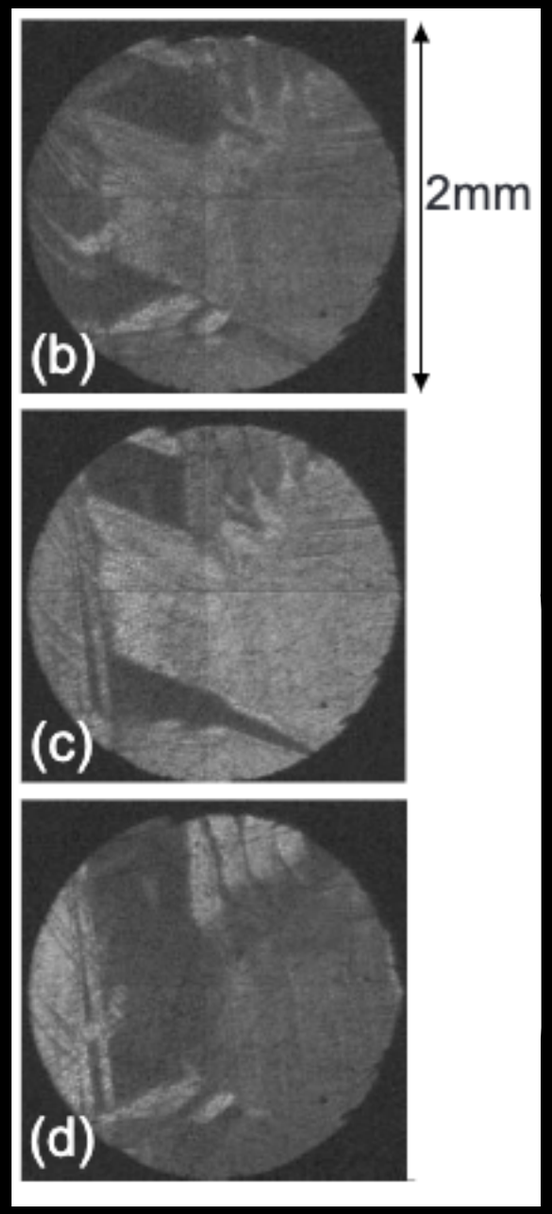
H. Gomonay and **D. Bossini**
J. Phys. D App. Phys 54, 374004 (2021)

Control experiment

✓ Repeat the experiment in a single-domain

Control experiment

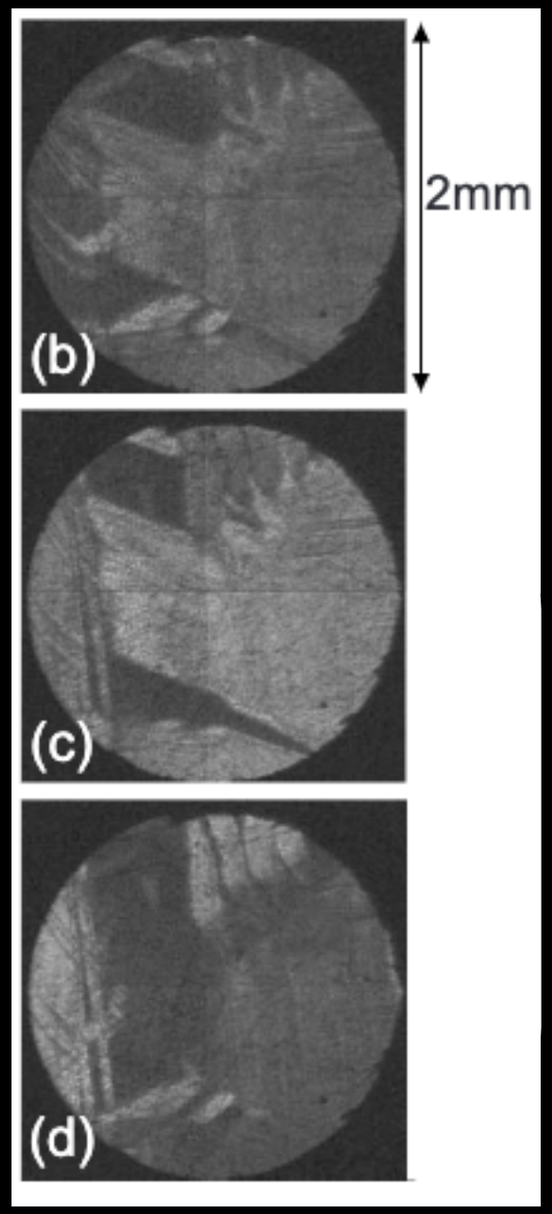
✓ Repeat the experiment in a single-domain



T. Satoh et al J. Opt. Soc. Am. B **7**, 1421 (2010)

Control experiment

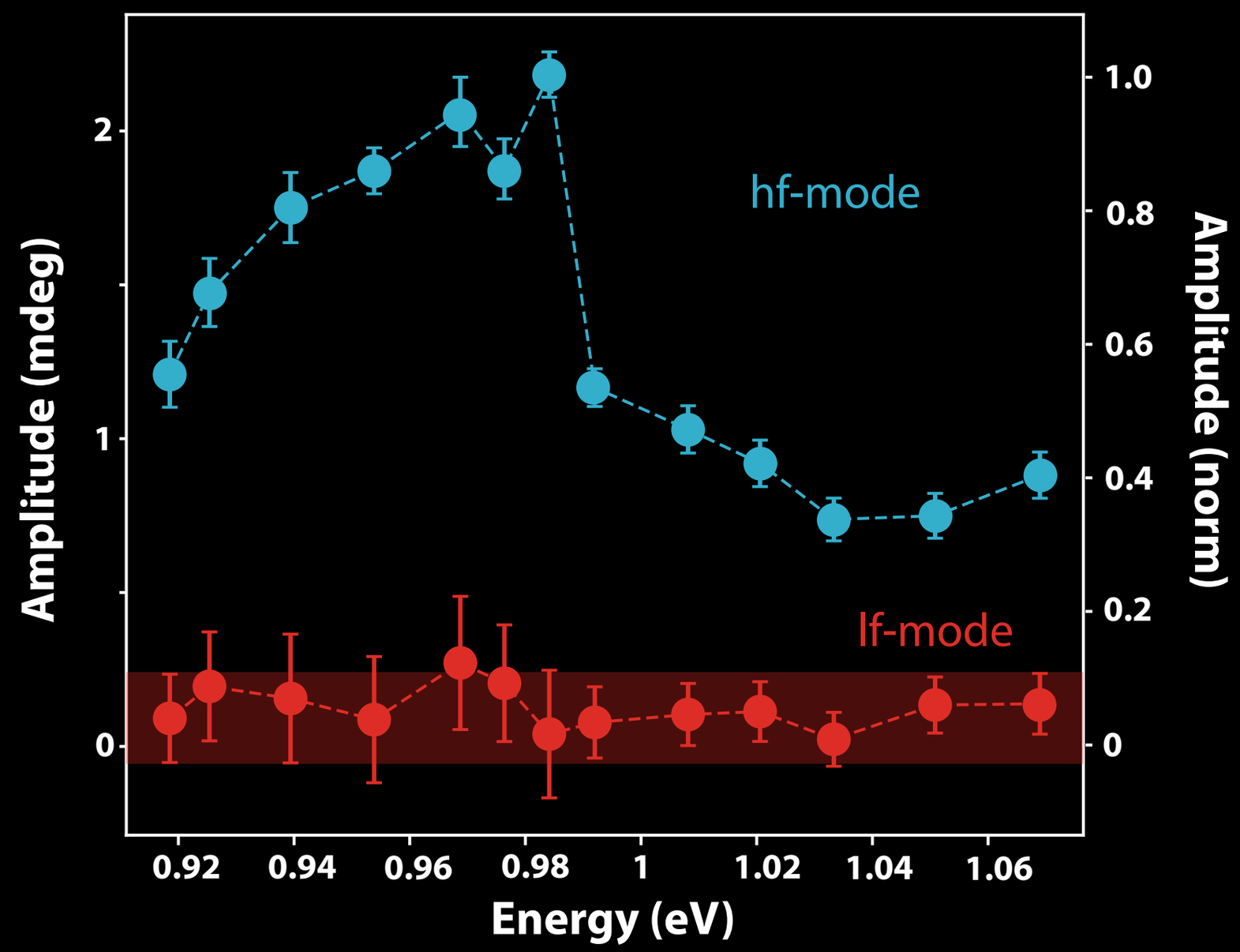
- ✓ Repeat the experiment in a single-domain
- ✓ Ar-O environment annealing (1400 °C)
- ✓ Domains > 100 micron



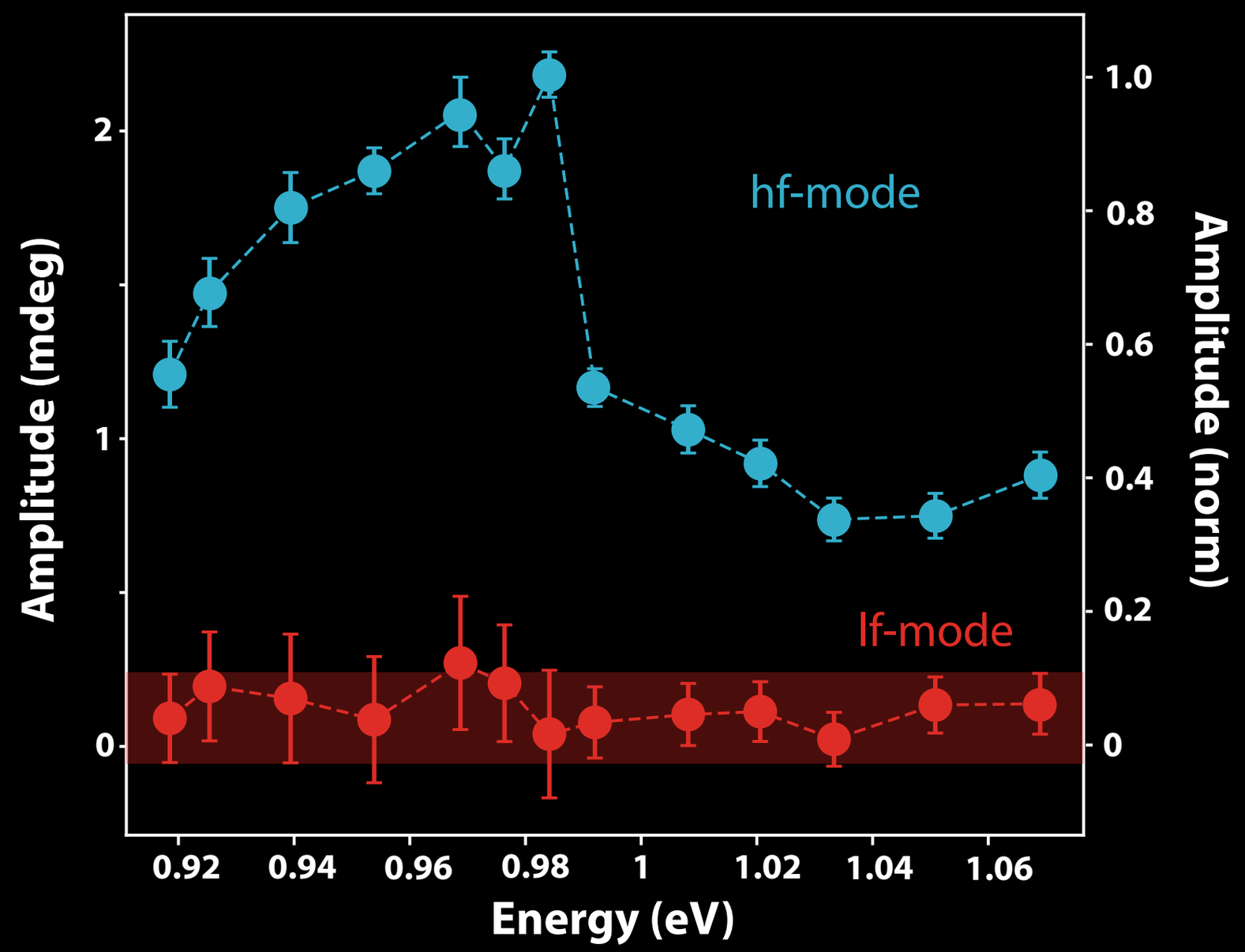
T. Satoh et al J. Opt. Soc. Am. B **7**, 1421 (2010)

Control experiment

Control experiment

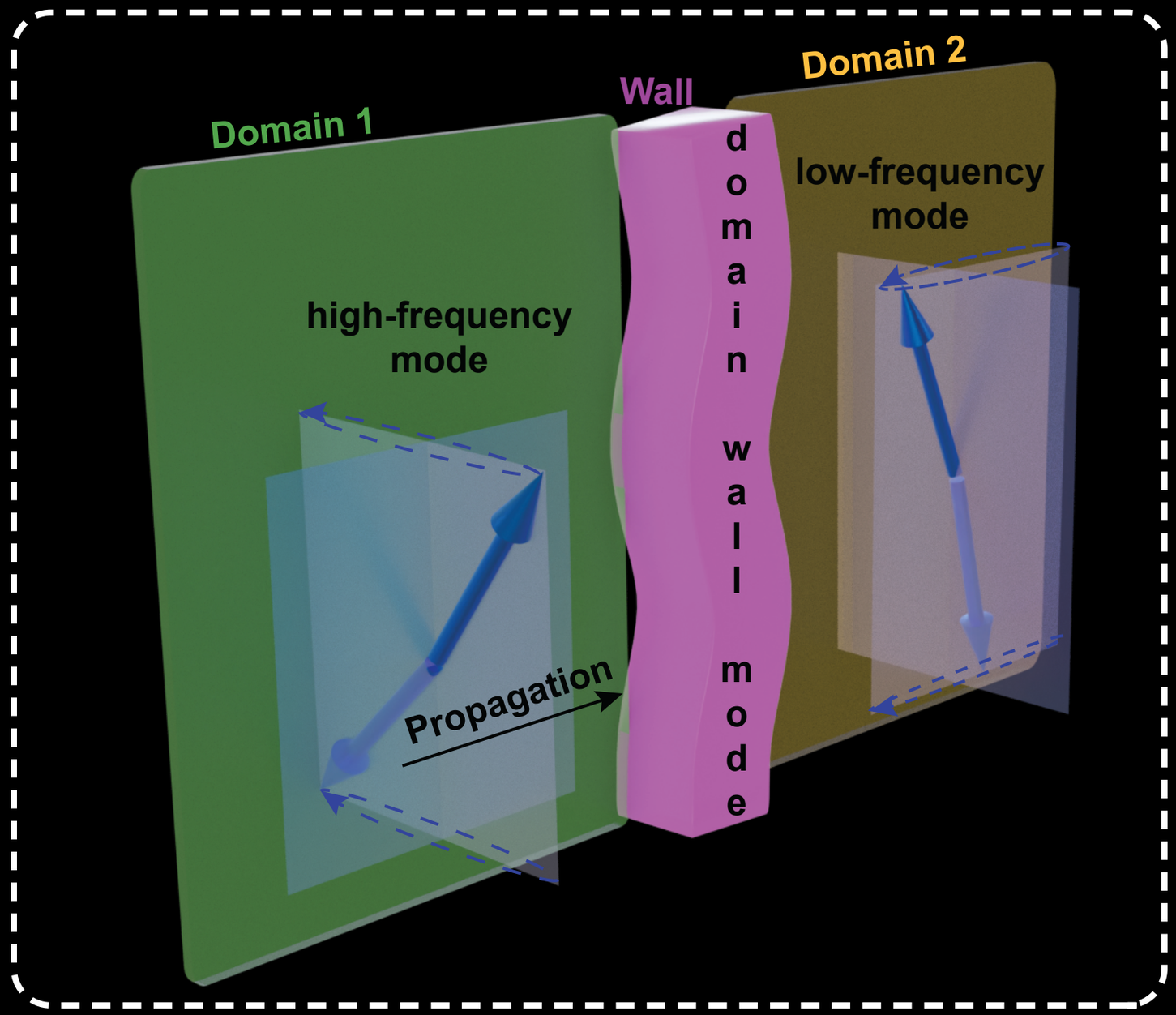


Control experiment



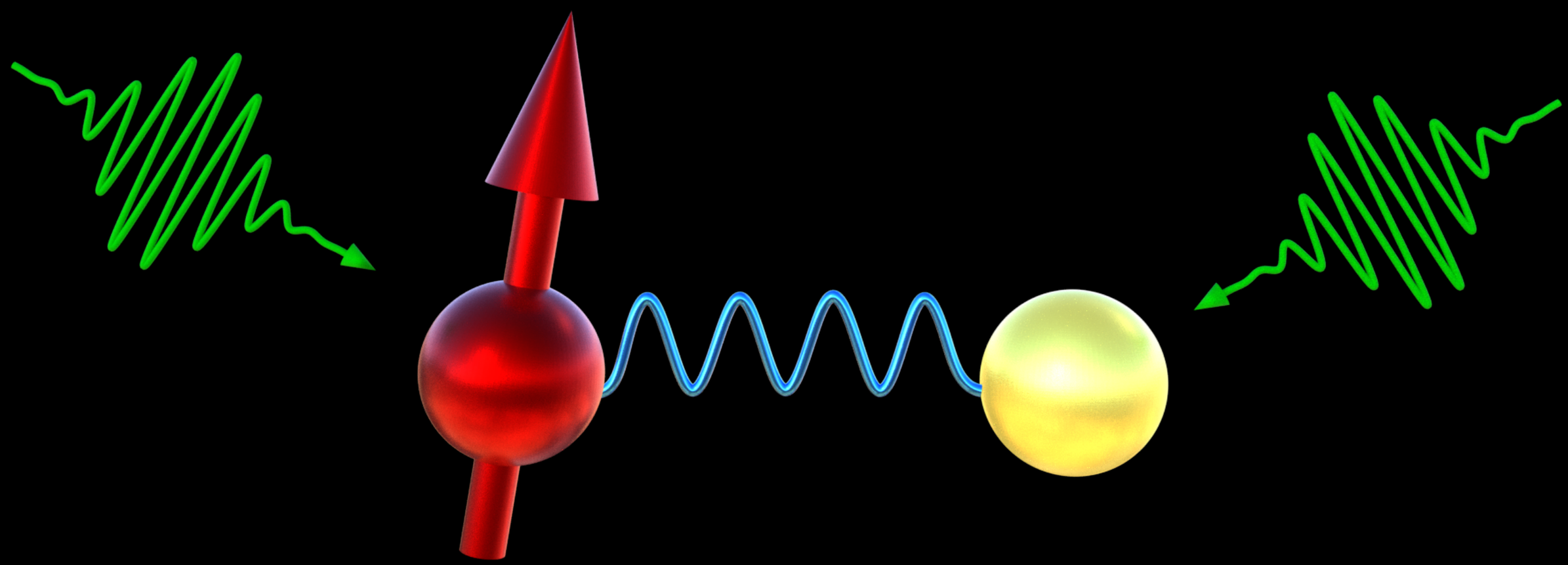
D. Bossini et al PRL **127**, 077202 (2021)

Conclusion

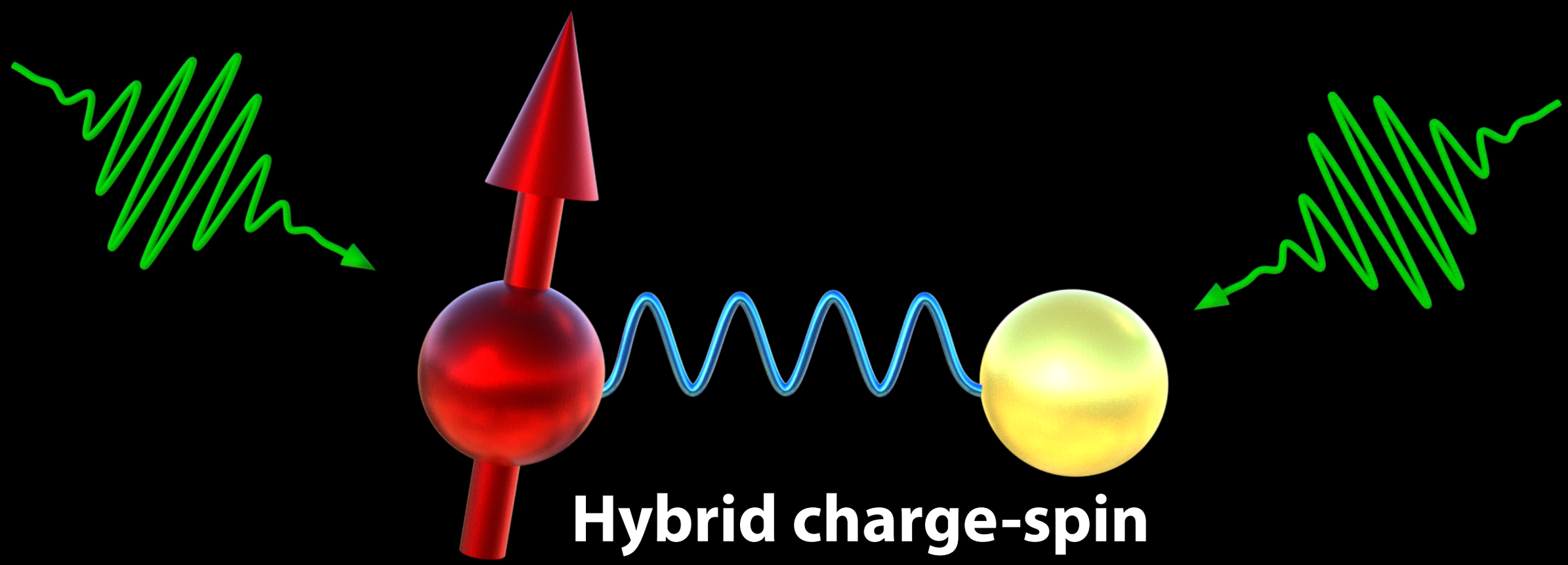


Walls-enabled nonlinear spin dynamics

Conclusion



Conclusion



Hybrid charge-spin excitation (X-M)

Photoinduced phase transition (600 fs)

Amplification magnon modes

Acknowledgements

Tokyo University

K. Konishi
T. Arima
S. Toyoda
M. Kuwata-Gonokami
E. Saitoh

Stockholm University

S. Bonetti
M. Pancaldi
M. Basini
L. Soumah

Mainz University

H. Gomonay
J. Sinova

TU Dortmund

F. Mertens
M. Cinchetti
G. Uhrig
M. Torbati
K. Deltenre

cost
EUROPEAN COOPERATION IN SCIENCE & TECHNOLOGY

Politecnico of Milano

S. Dal Conte
G. Cerullo

Tokyo Institute of Technology

T. Satoh

Emmy Noether-Programm
DFG Deutsche Forschungsgemeinschaft

#StandwithUkraine