Femtosecond quantum spin dynamics in antiferromagnets

D. Bossini

JSPS "Overseas Researcher" Fellow at University of Tokyo, Japan

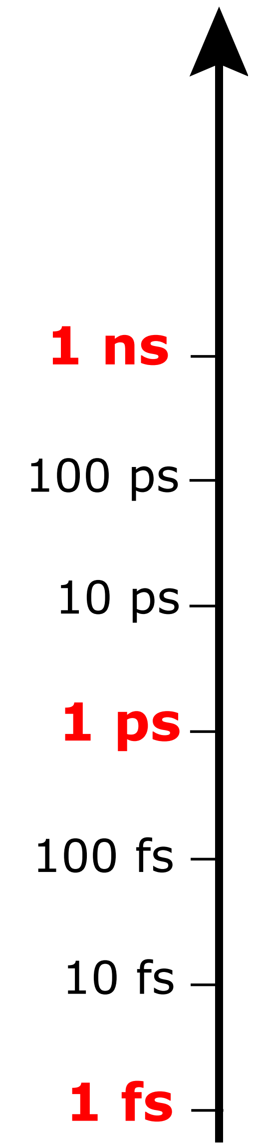
Scientific goal

Scientific goal

Ultrafast manipulation of the magnetic order

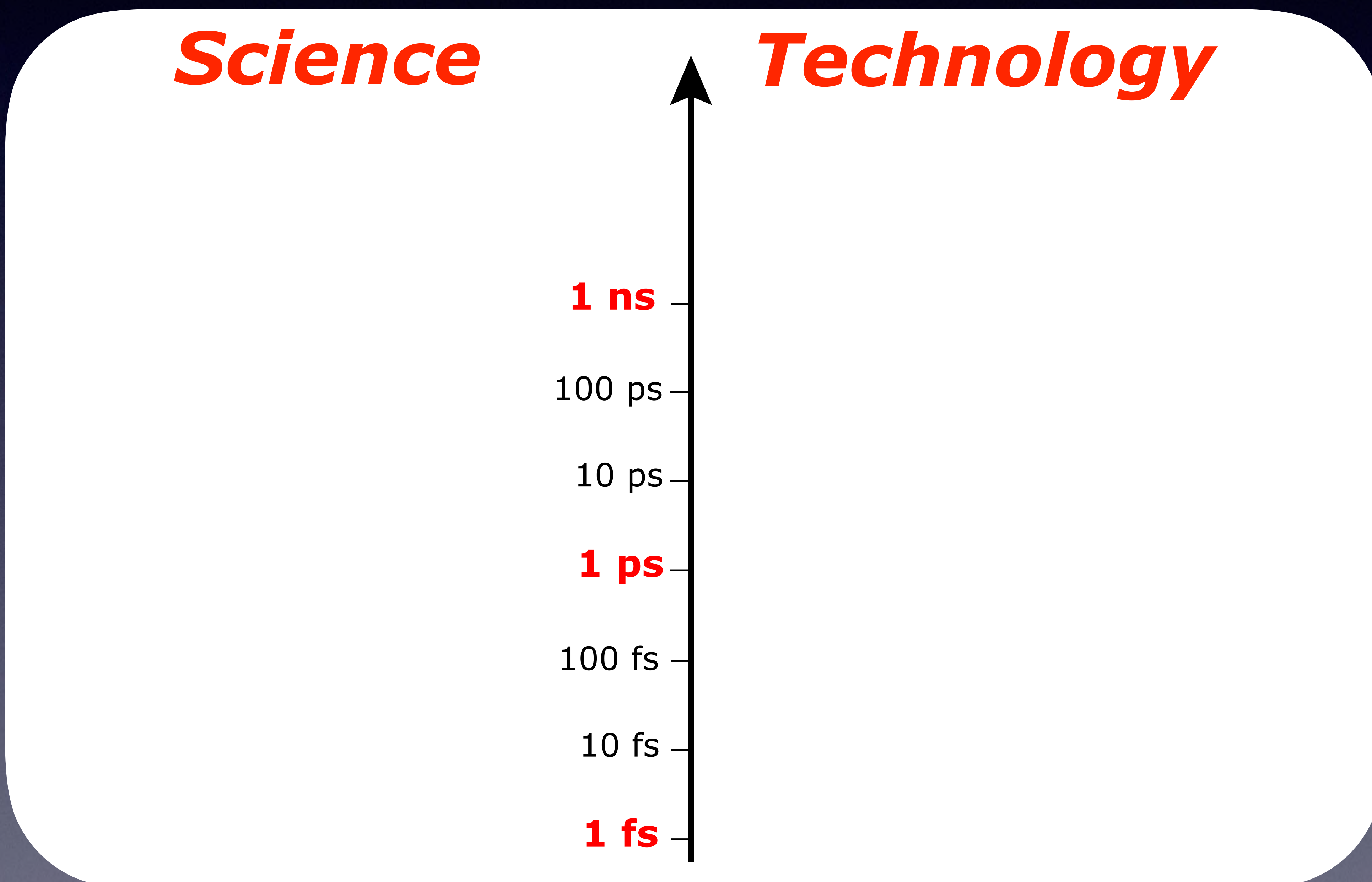
Scientific goal

Ultrafast manipulation of the magnetic order



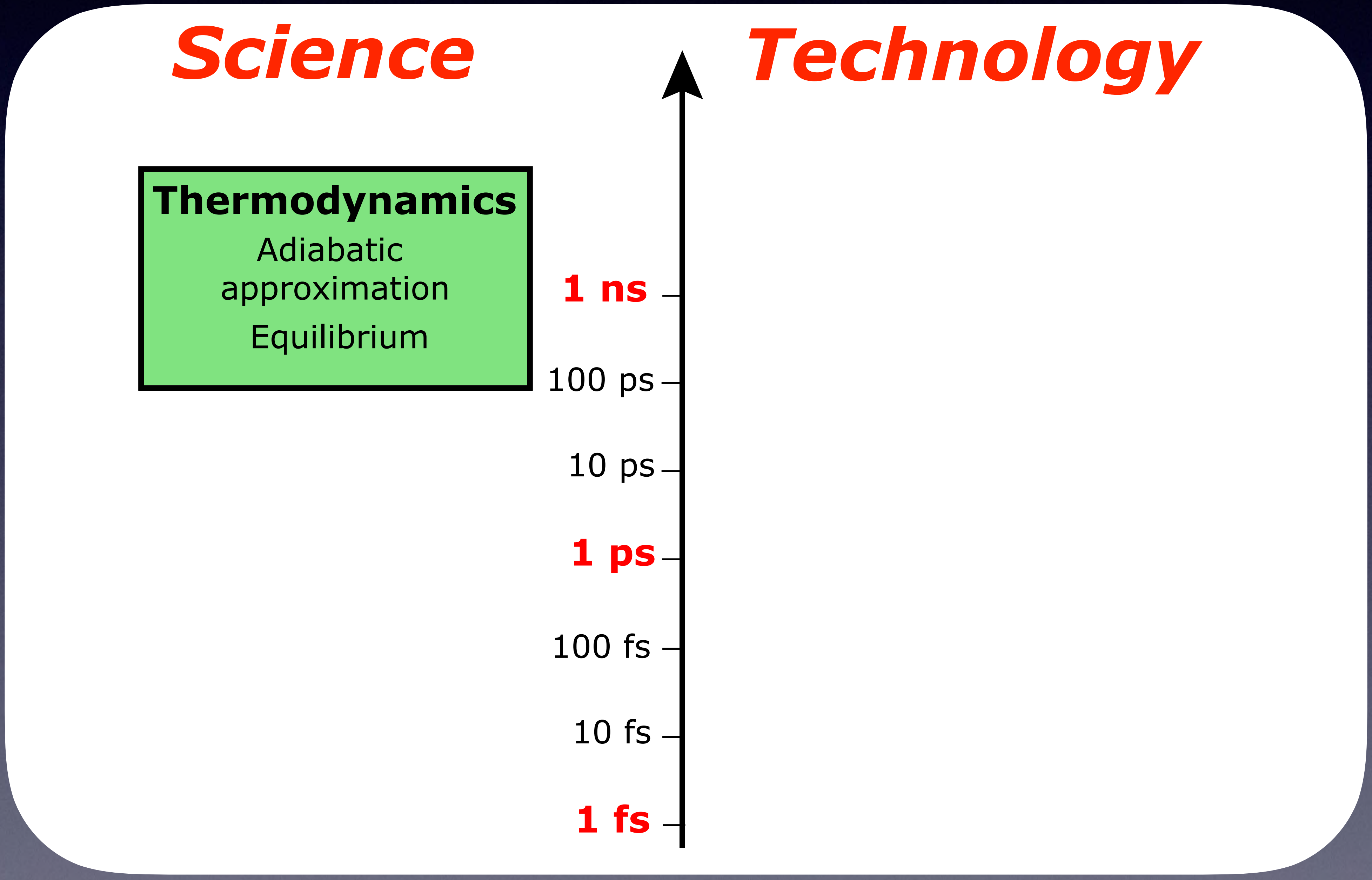
Scientific goal

Ultrafast manipulation of the magnetic order



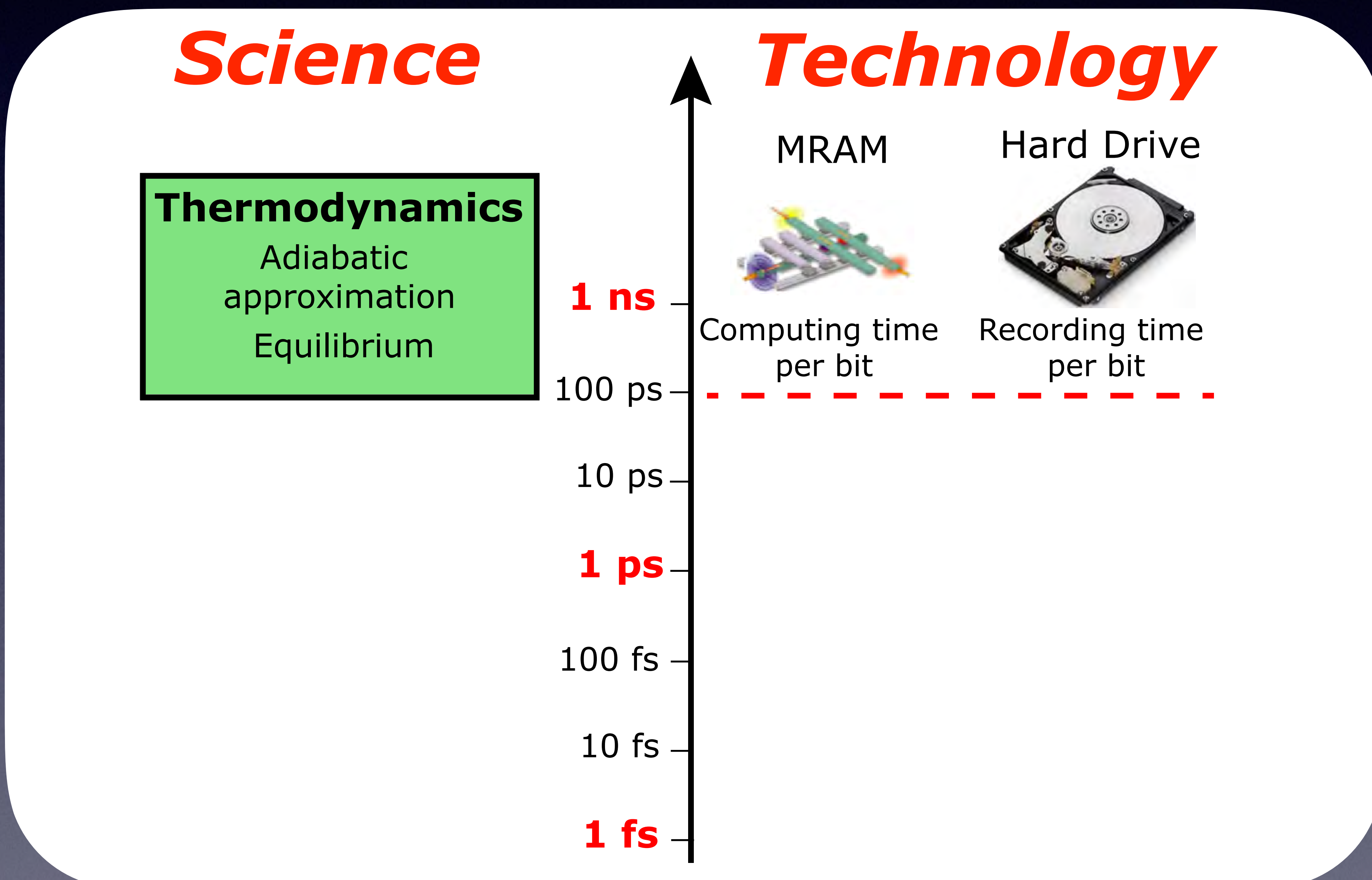
Scientific goal

Ultrafast manipulation of the magnetic order



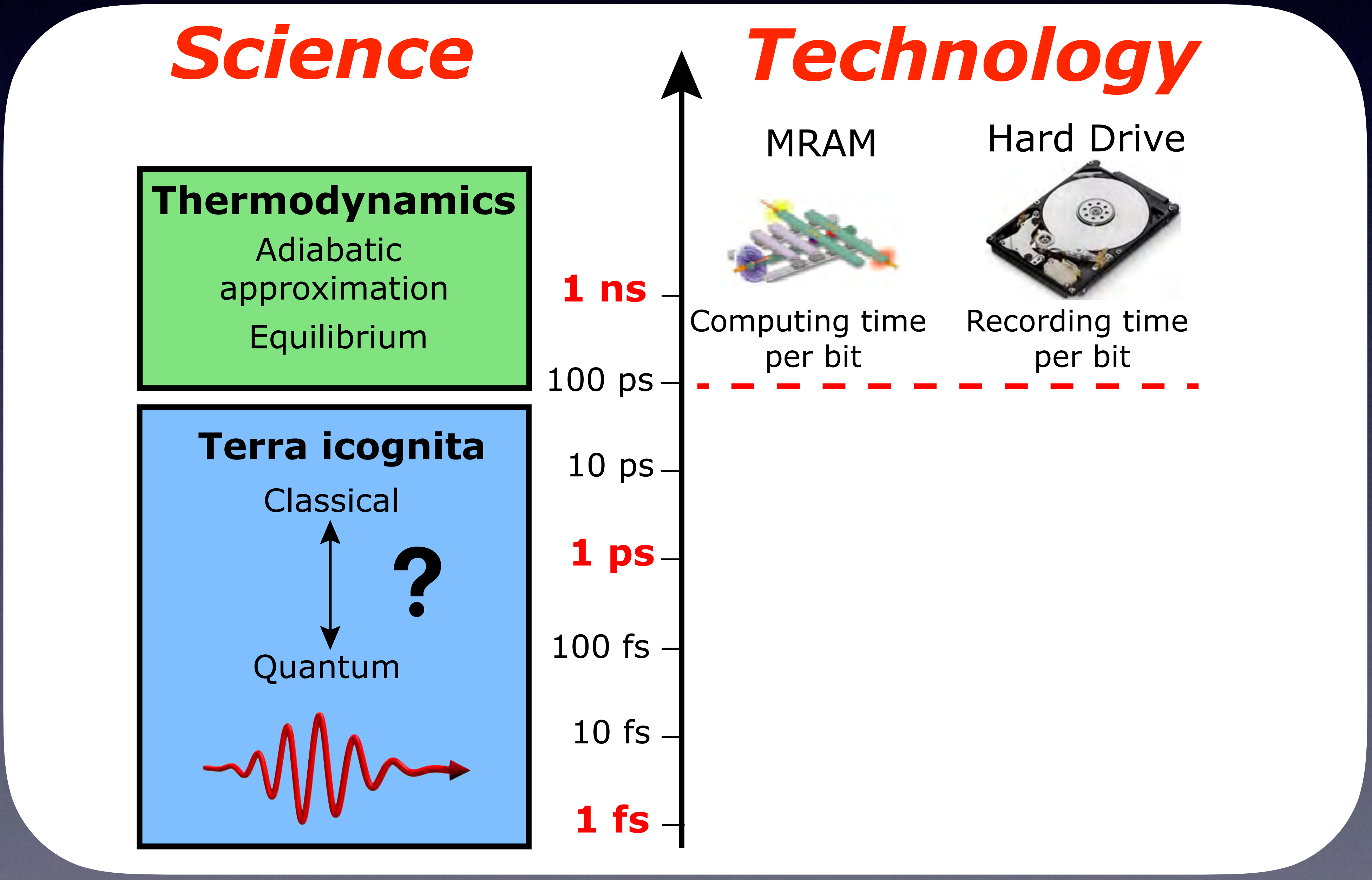
Scientific goal

Ultrafast manipulation of the magnetic order



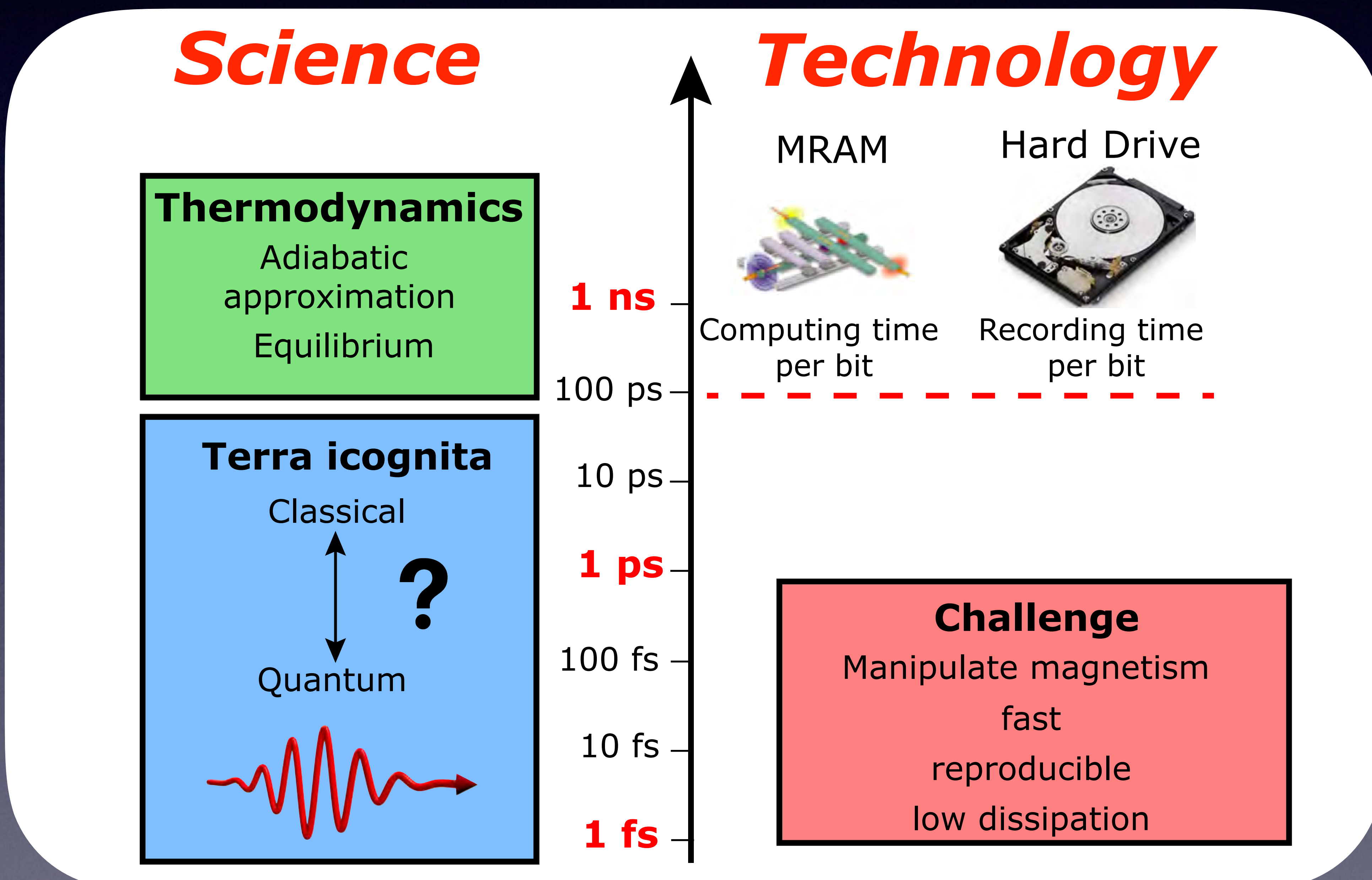
Scientific goal

Ultrafast manipulation of the magnetic order



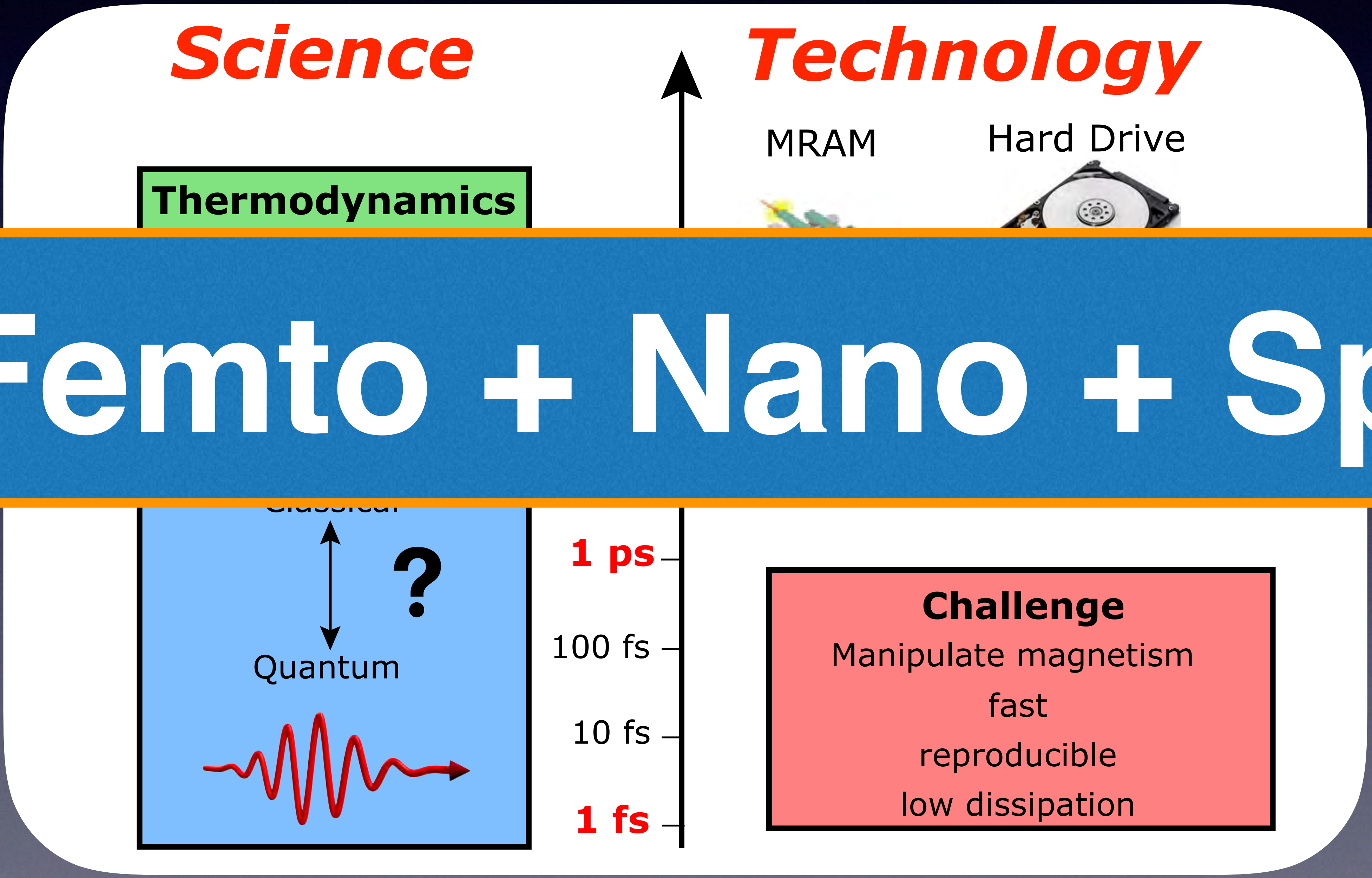
Scientific goal

Ultrafast manipulation of the magnetic order



Scientific goal

Ultrafast manipulation of the magnetic order



Femto + Nano + Spin

Dielectric antiferromagnet

Dielectric antiferromagnet

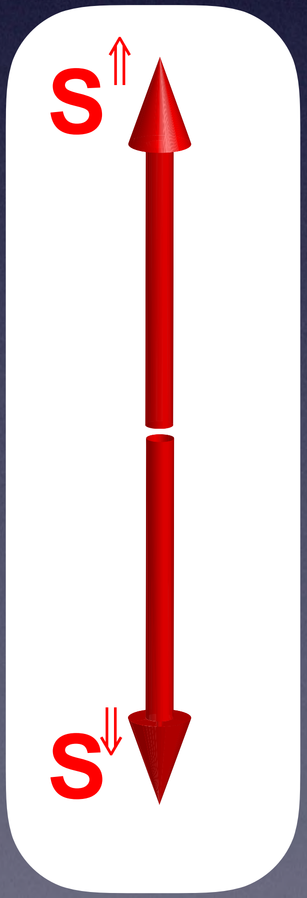
- ✓ No free electrons
- ✓ Majority of magnetically ordered materials
- ✓ No stray field, technological potential
- ✓ Intrinsically faster spin dynamics

Dielectric antiferromagnet

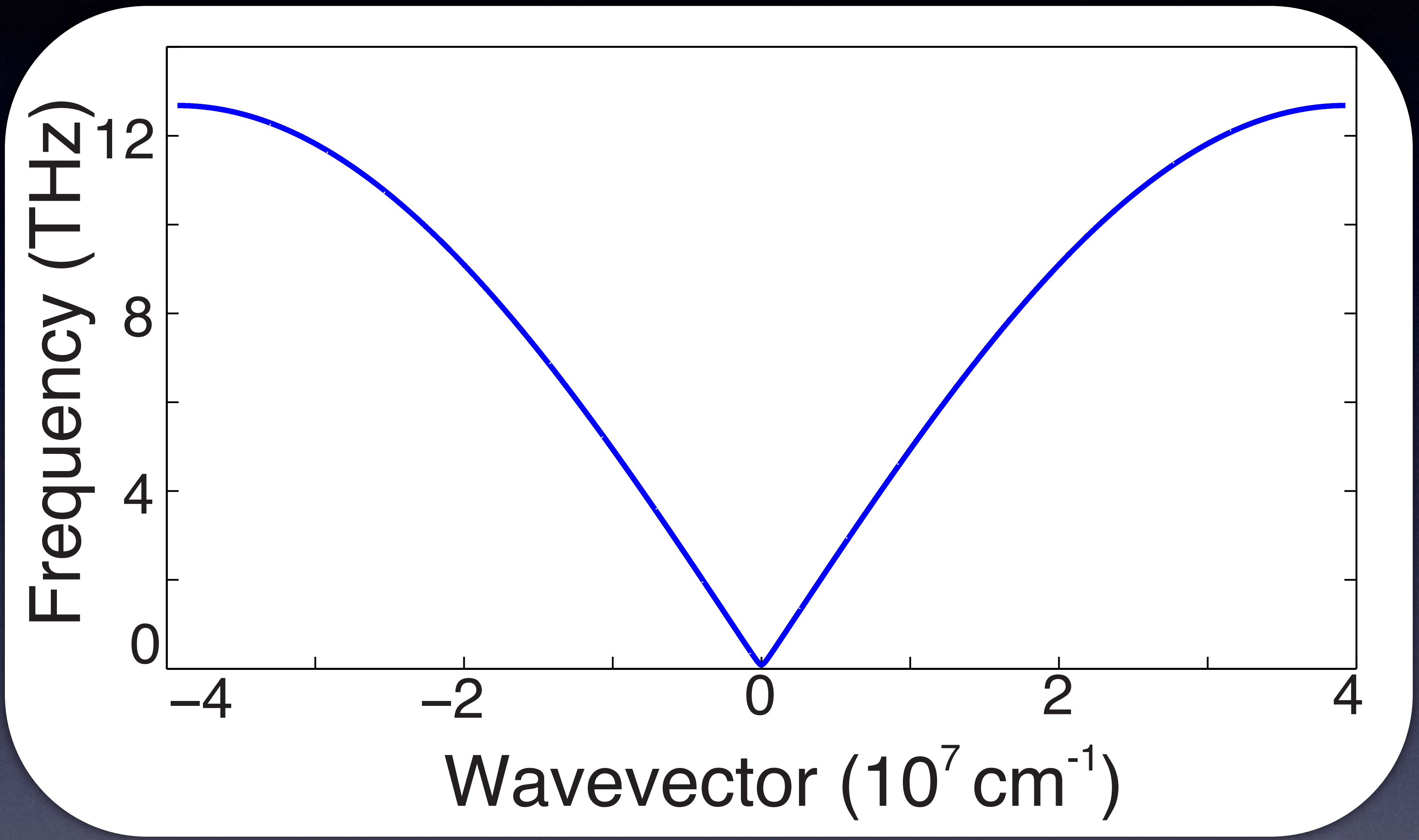
- ✓ No free electrons
- ✓ Majority of magnetically ordered materials
- ✓ No stray field, technological potential
- ✓ Intrinsically faster spin dynamics

$$\hat{H} = J \sum_{\langle i,j \rangle} \hat{\boldsymbol{S}}_i \cdot \hat{\boldsymbol{S}}_j$$

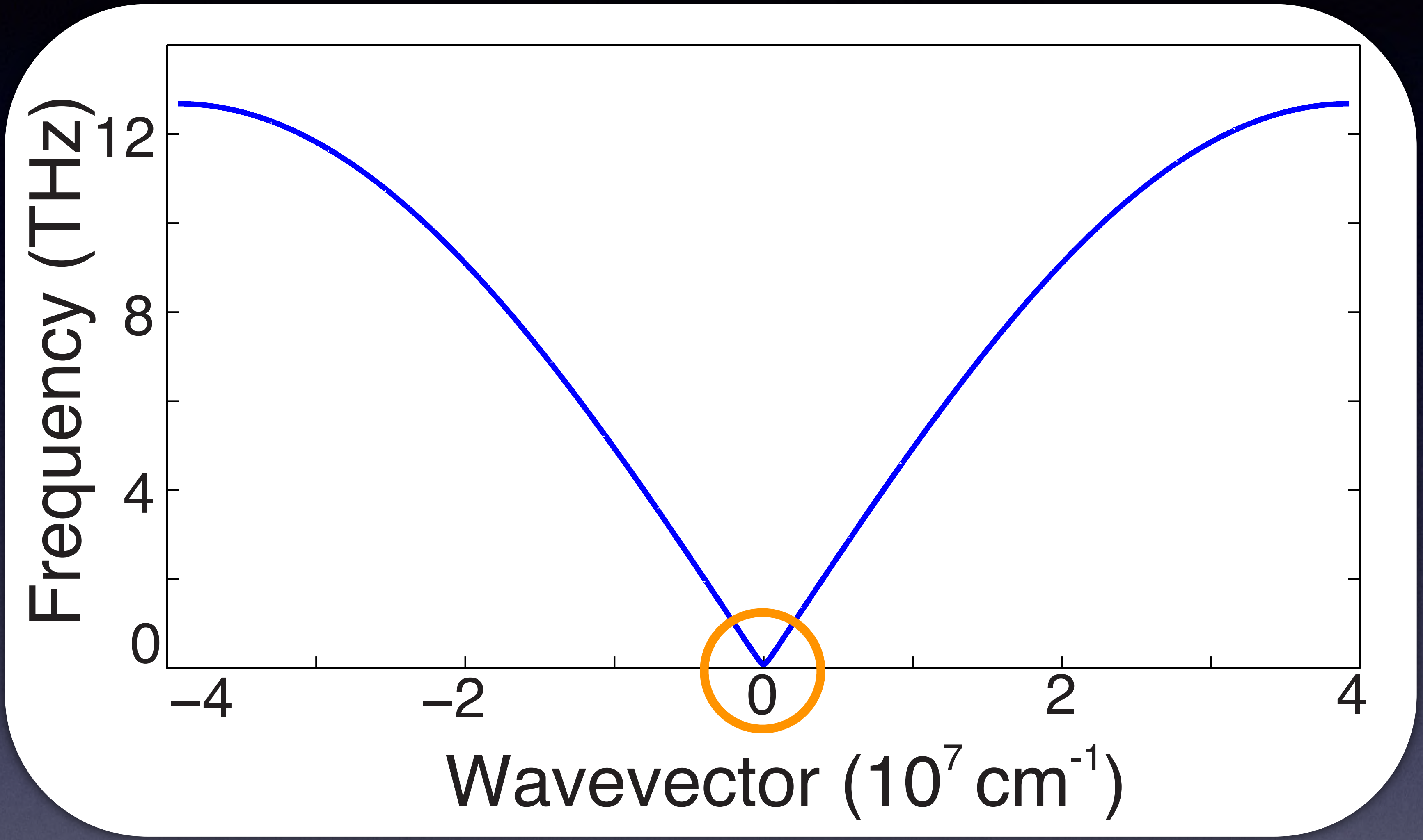
Collinear magnetic sublattices



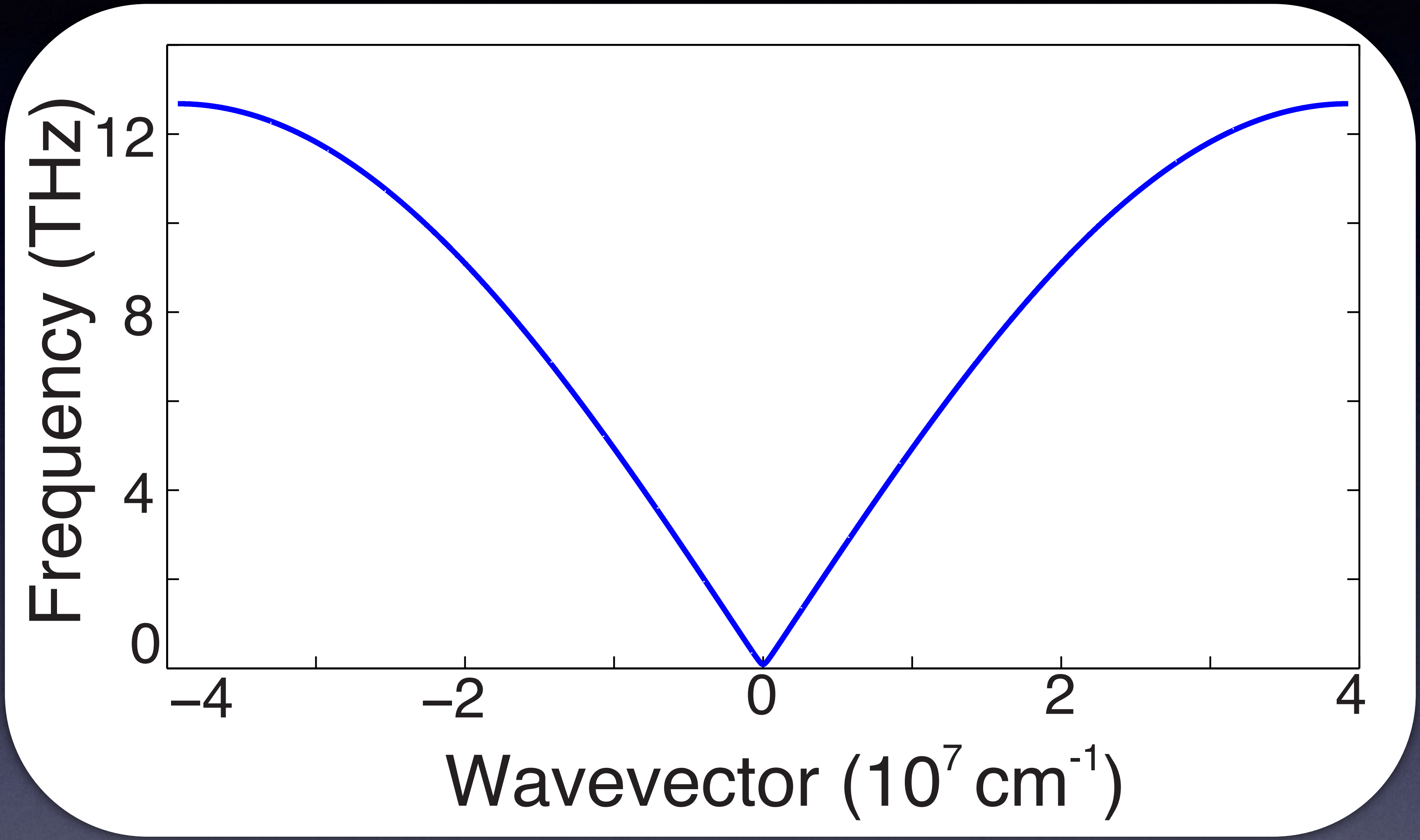
Femto-nanomagnonics



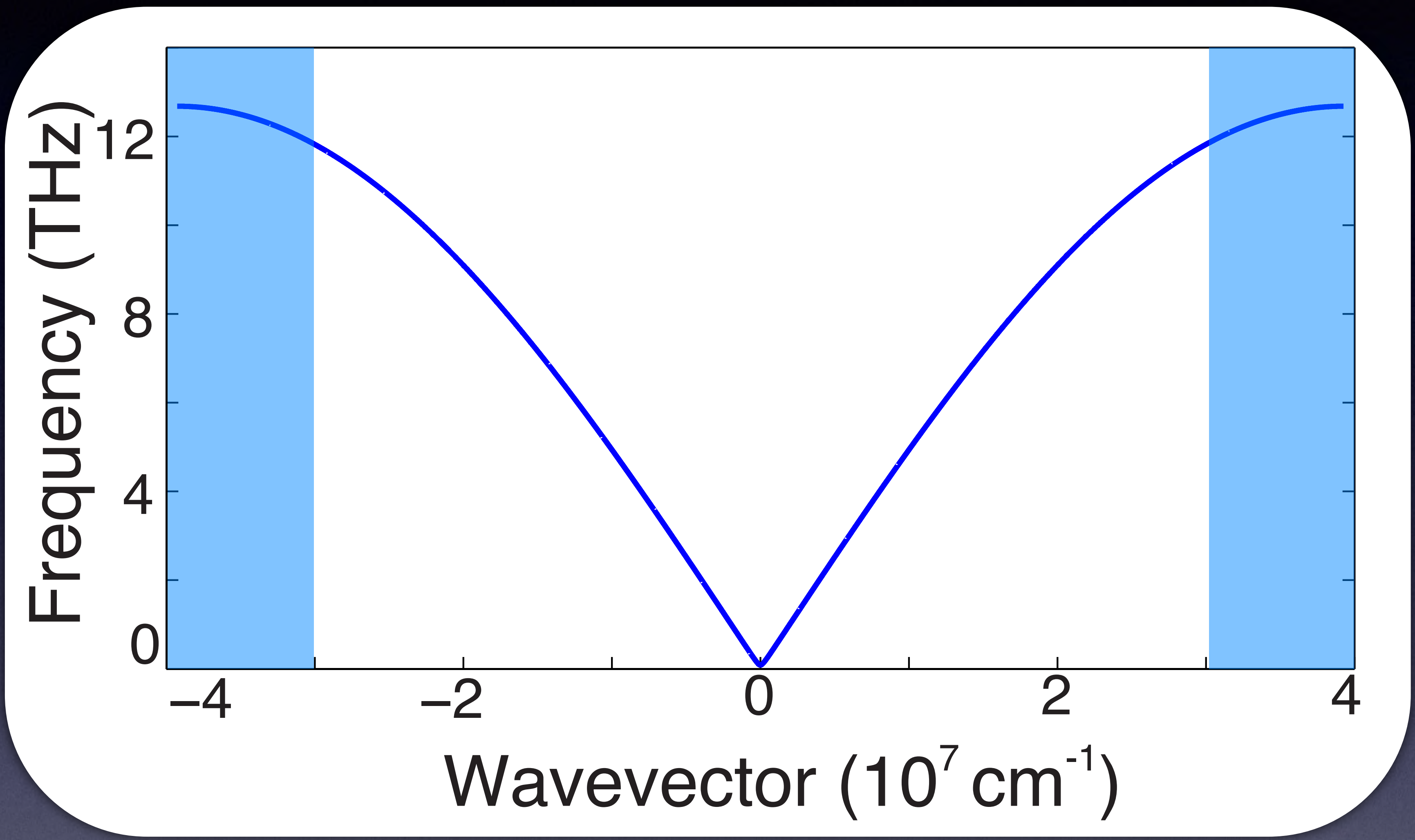
Femto-nanomagnonics



Femto-nanomagnonics

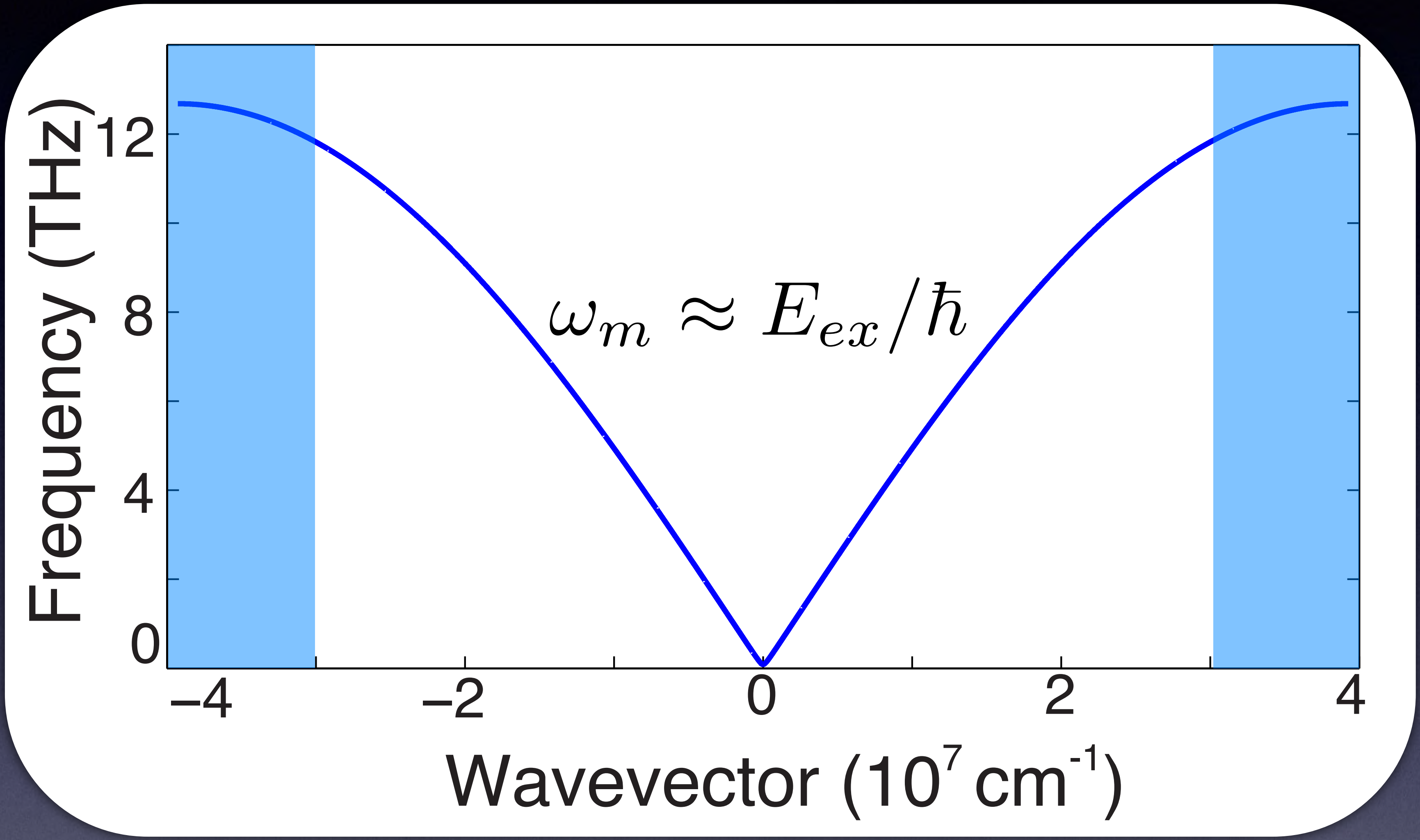


Femto-nanomagnonics



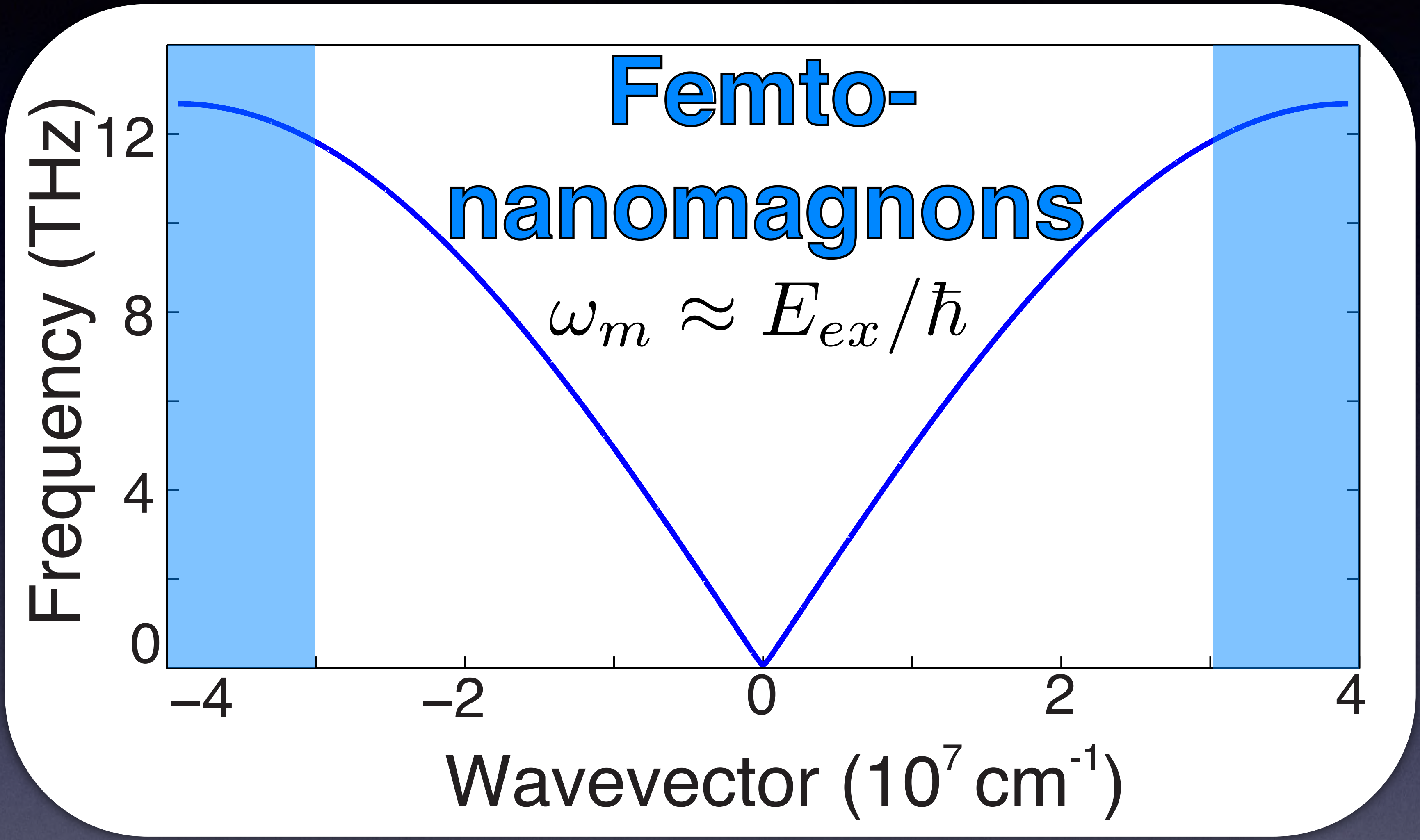
- ✓ Femtosecond period
- ✓ Nanometer wavelength
- ✓ Defined by E_{ex}

Femto-nanomagnonics



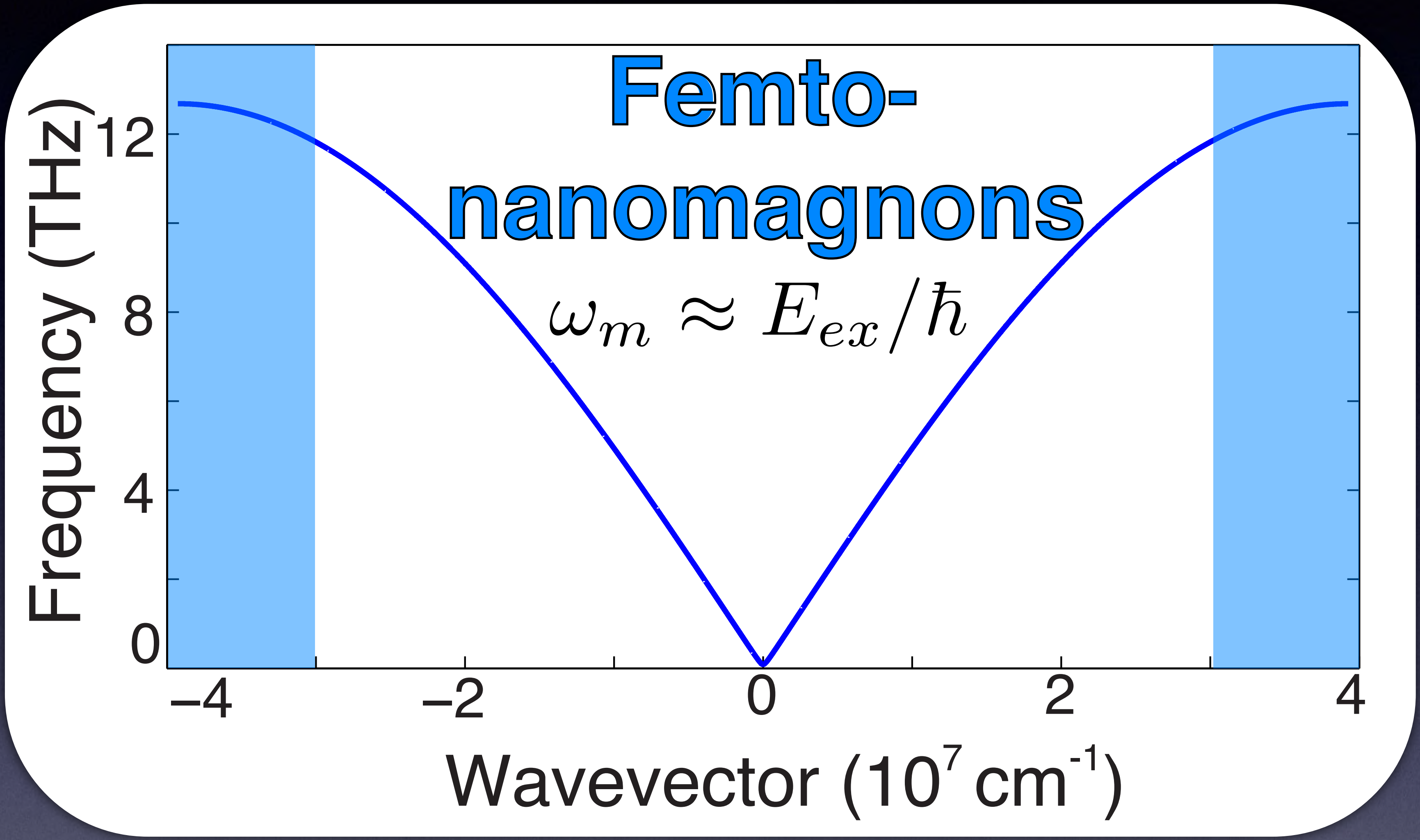
- ✓ Femtosecond period
- ✓ Nanometer wavelength
- ✓ Defined by E_{ex}

Femto-nanomagnonics



- ✓ Femtosecond period
- ✓ Nanometer wavelength
- ✓ Defined by E_{ex}

Femto-nanomagnonics



- ✓ Femtosecond period
- ✓ Nanometer wavelength
- ✓ Defined by E_{ex}

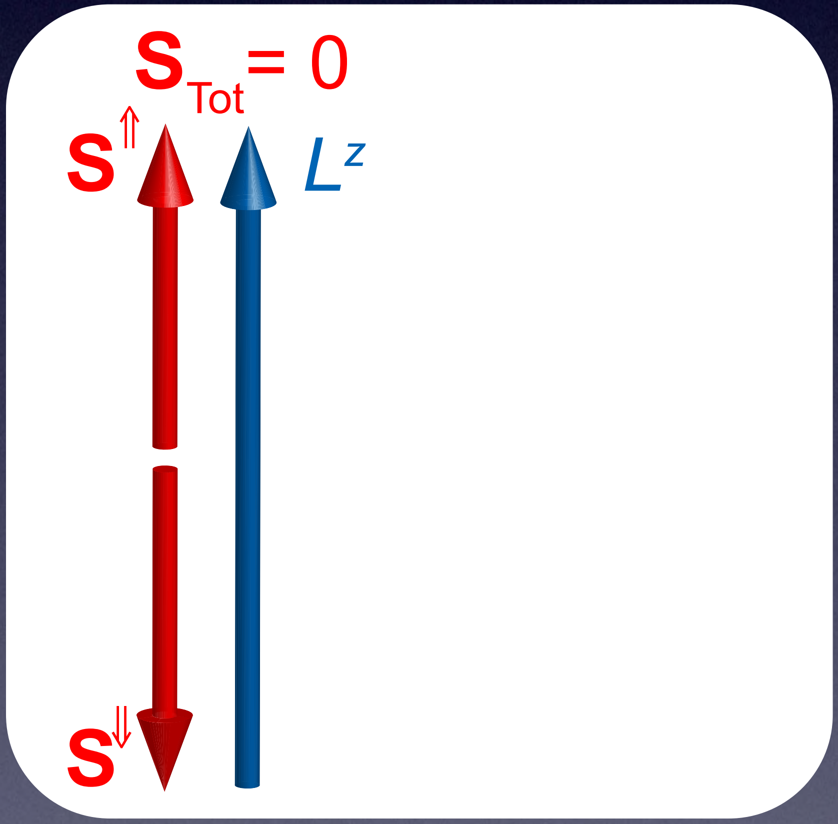
Measure spin dynamics triggered by femto-nanomagnons

Generation

Problem: high-wavevector magnons are usually unaccessible

Generation

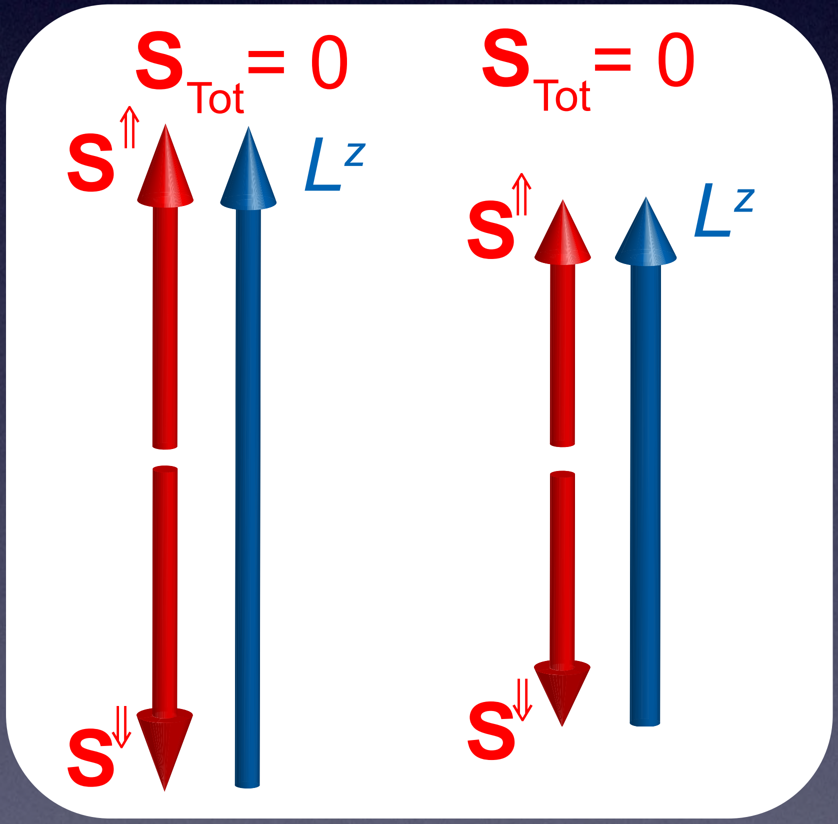
Problem: high-wavevector magnons are usually unaccessible



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

Generation

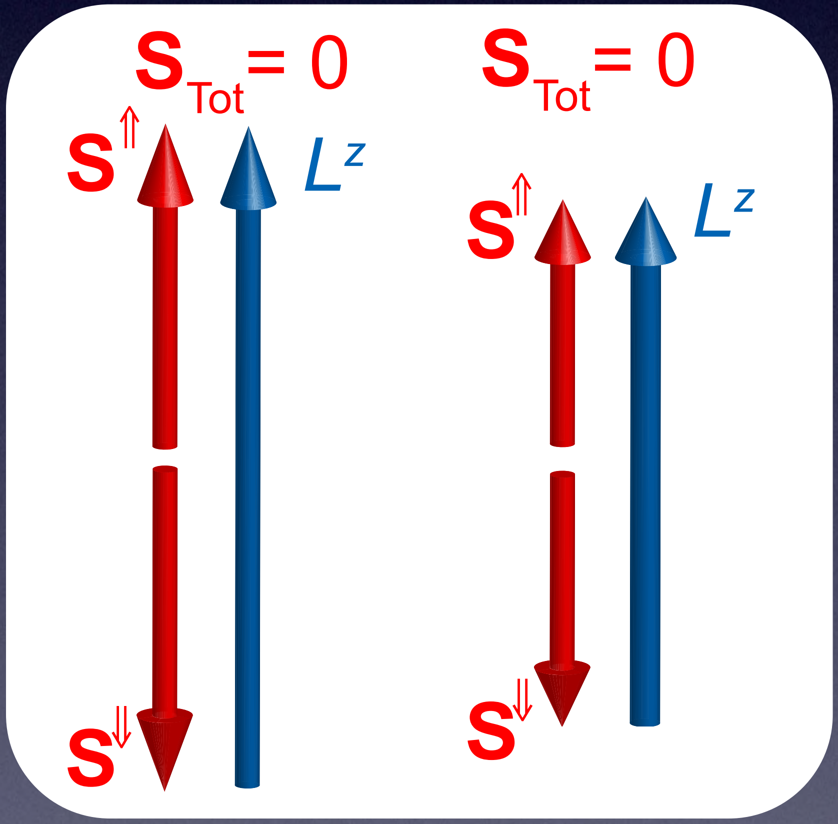
Problem: high-wavevector magnons are usually unaccessible



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

Generation

Problem: high-wavevector magnons are usually unaccessible

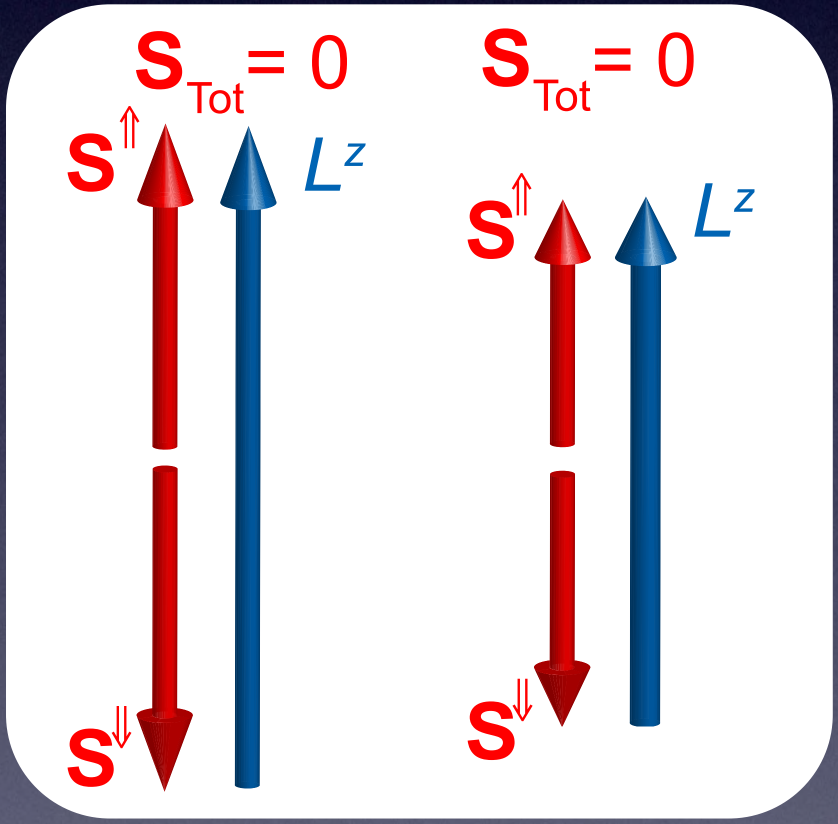


$$L \equiv S^{\Uparrow} - S^{\Downarrow}$$

- ✓ Spin and momentum conservation
- ✓ Light-induced bound state of a magnon pair: ***two-magnon mode***
- ✓ High-wavevector region: DOS

Generation

Problem: high-wavevector magnons are usually unaccessible



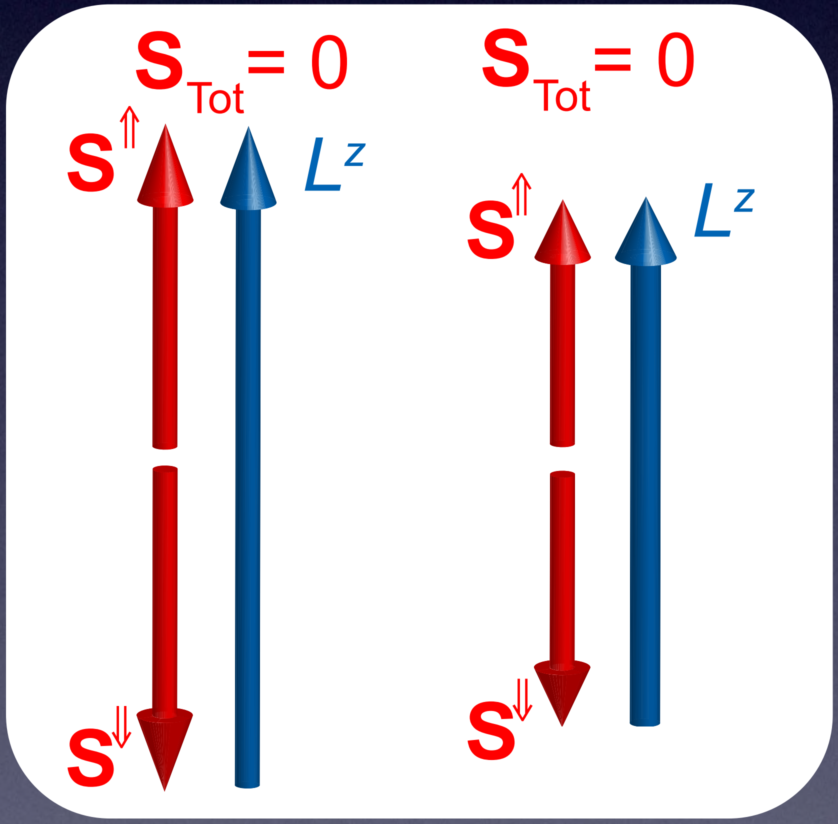
$$L \equiv S^{\Uparrow} - S^{\Downarrow}$$

- ✓ Spin and momentum conservation
- ✓ Light-induced bound state of a magnon pair: ***two-magnon mode***
- ✓ High-wavevector region: DOS

$$E_{2M} = E_{ex} + \Delta$$

Generation

Problem: high-wavevector magnons are usually unaccessible

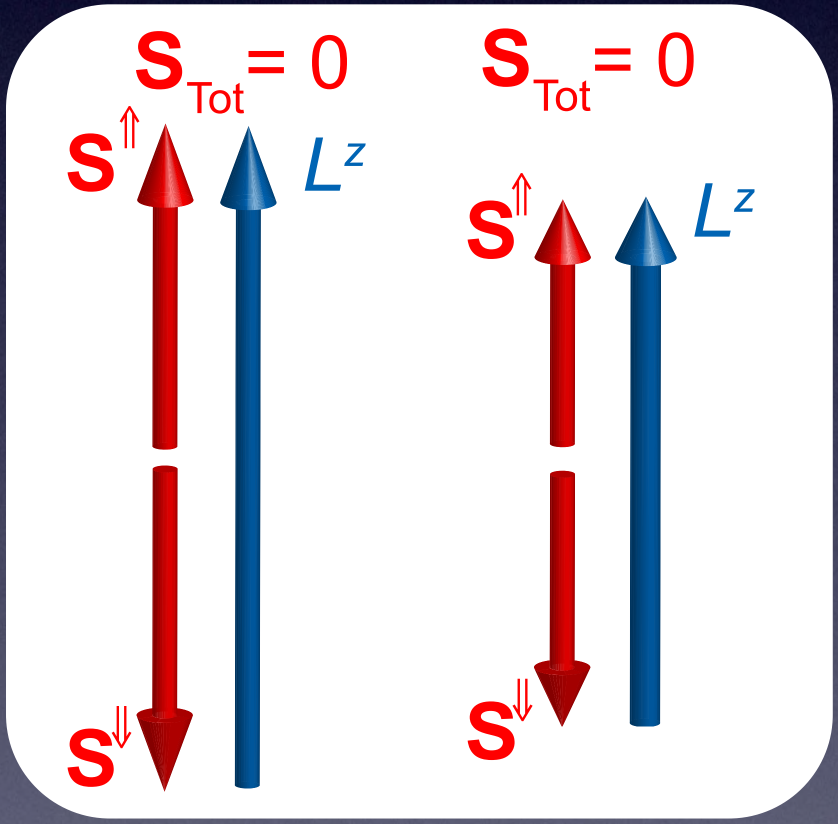


$$E_{2M} = E_{ex} + \Delta$$

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

Generation

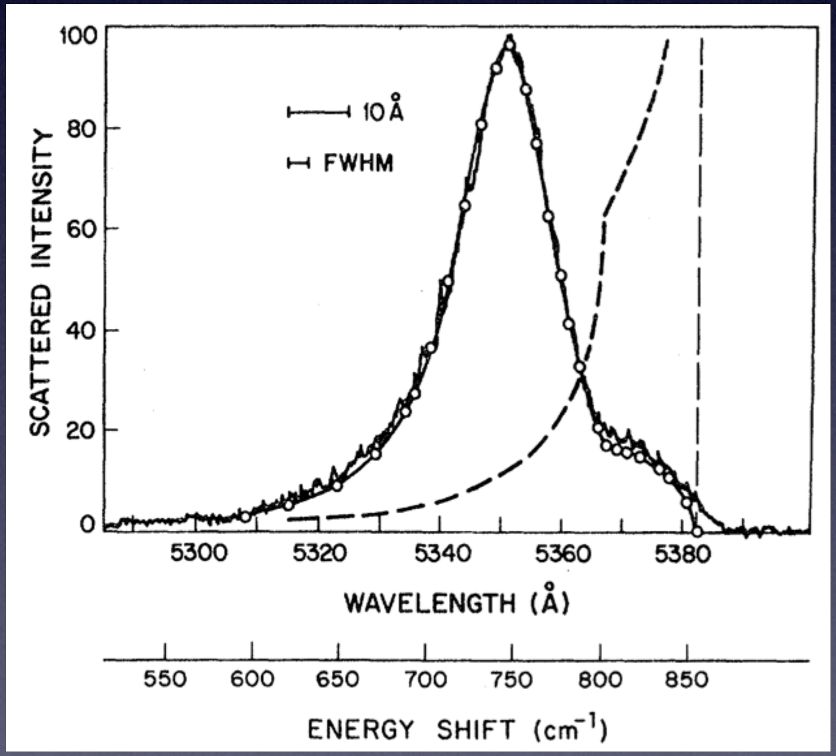
Problem: high-wavevector magnons are usually unaccessible



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

$KNiF_3$

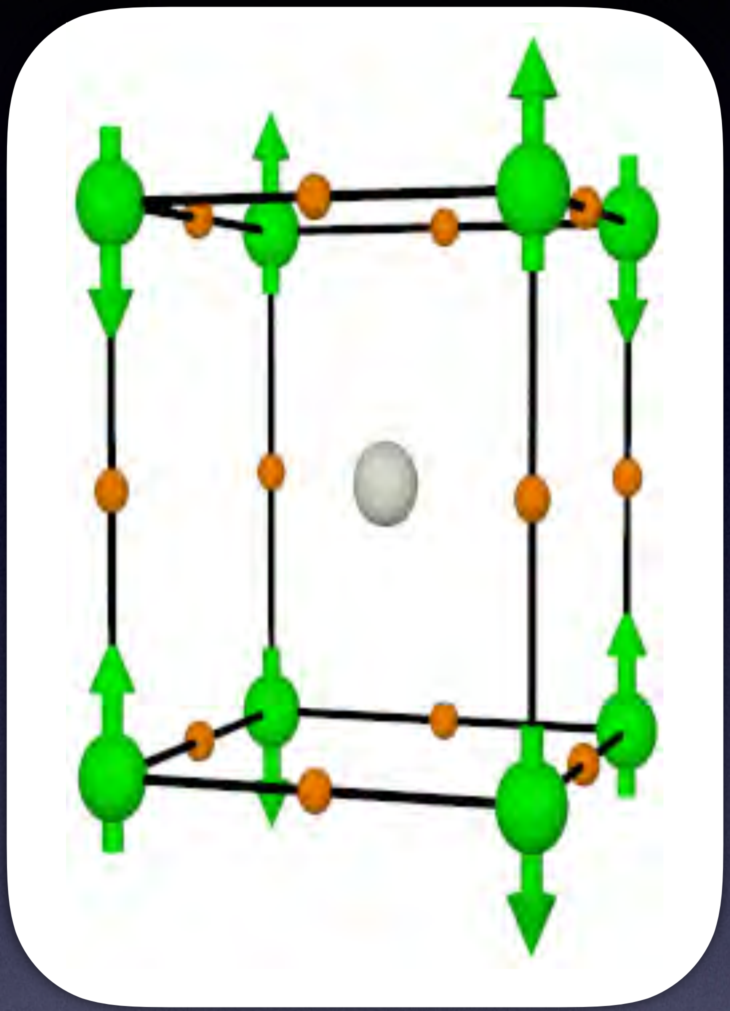
$$E_{2M} = E_{ex} + \Delta$$



S. Chinn et al. PRB **3**, 1709 (1971)

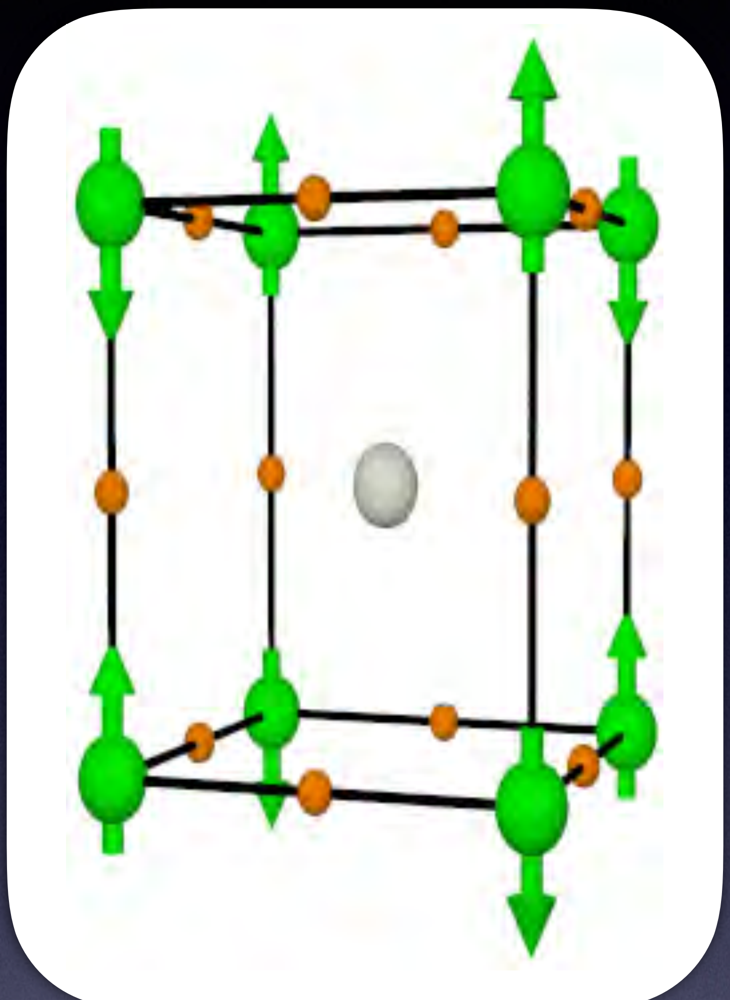
U. Balucani et al. PRB **8**, 4247 (1973)

Sample: $KNiF_3$

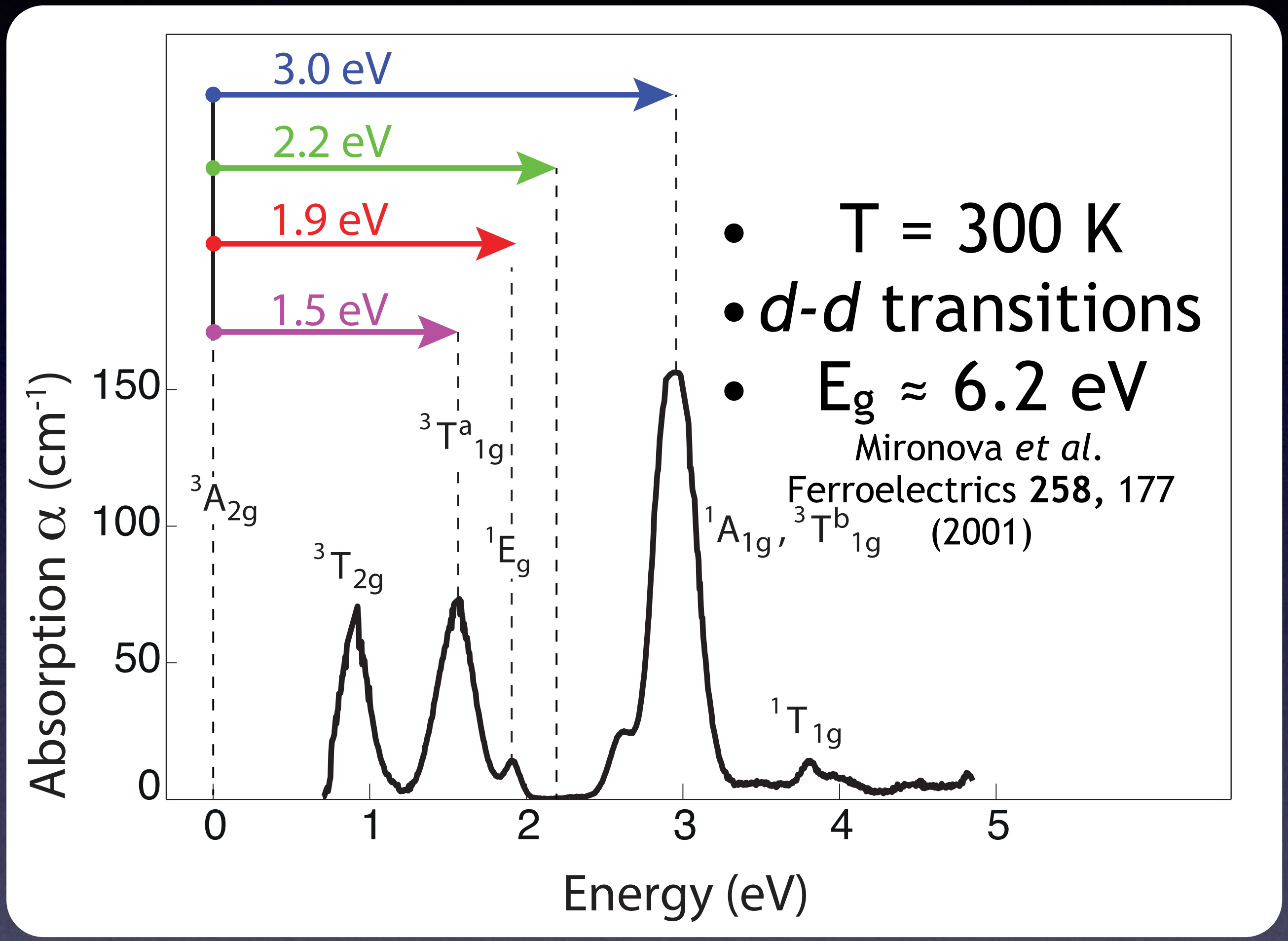


$T_N = 246$ K

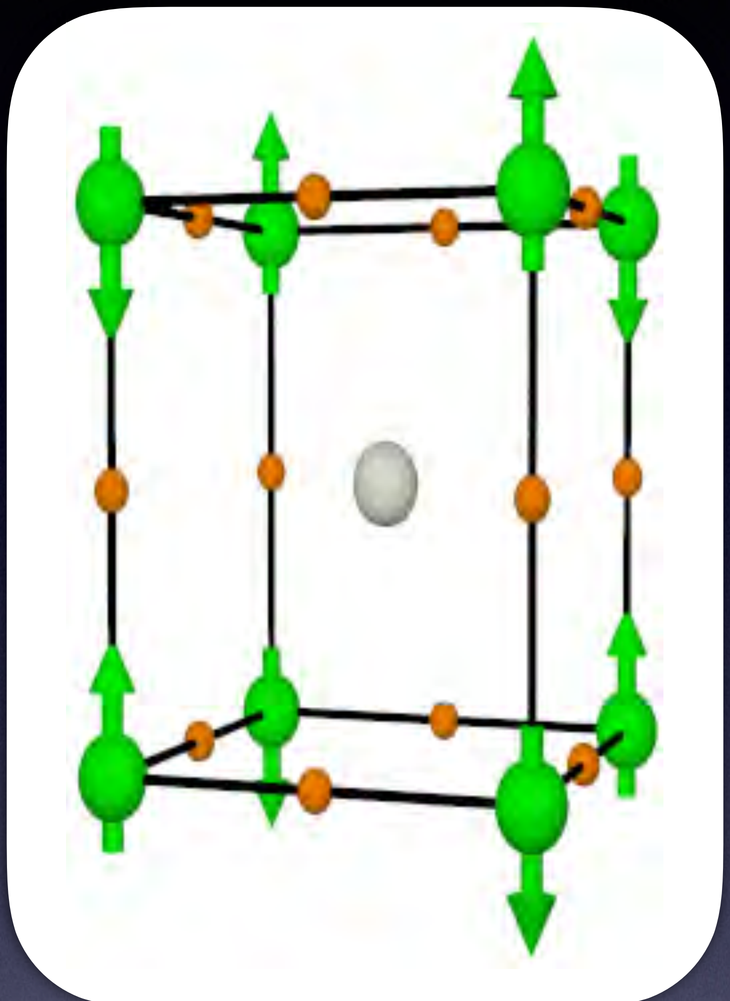
Sample: $KNiF_3$



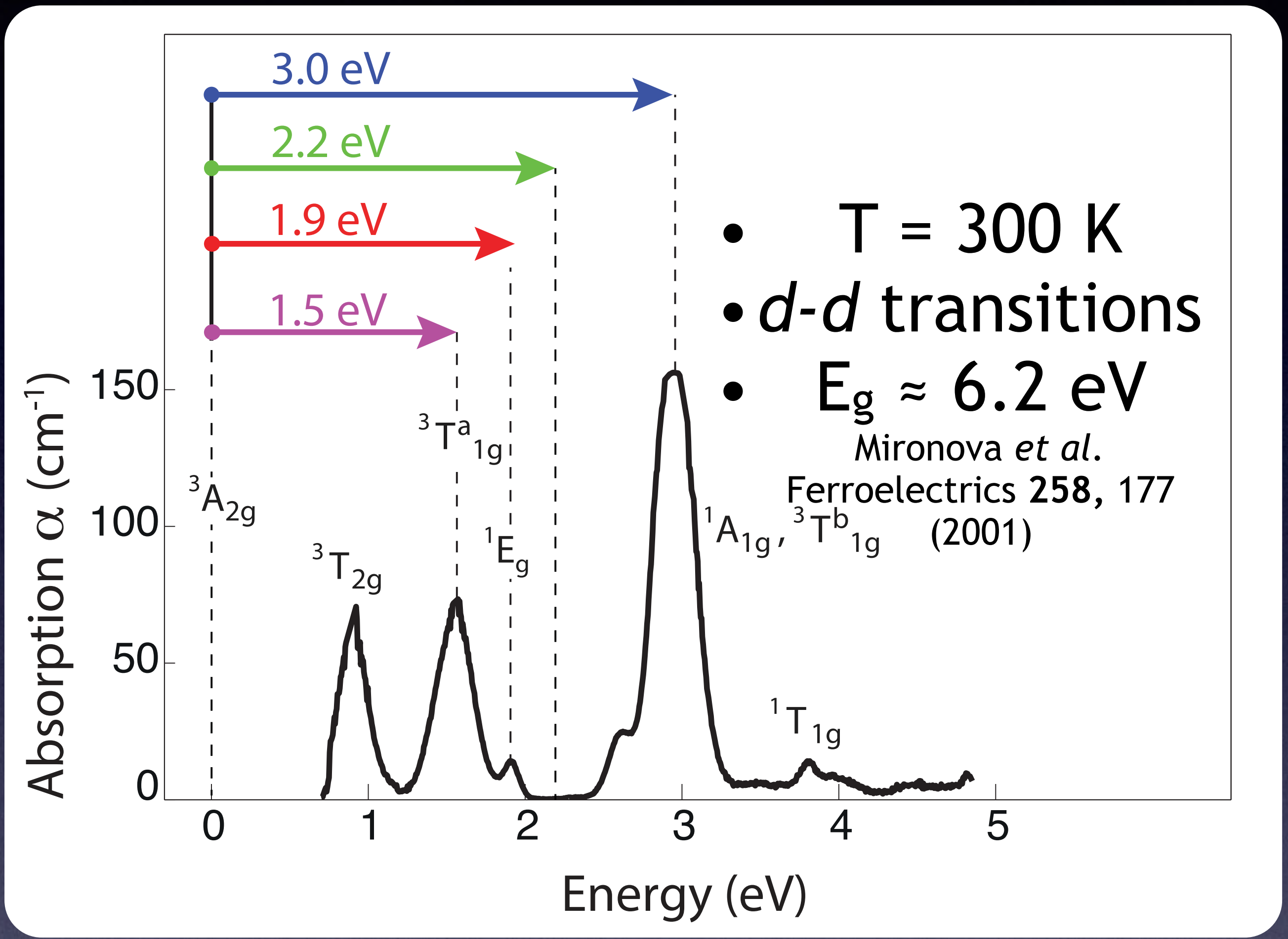
T_N = 246 K



Sample: $KNiF_3$



T_N = 246 K

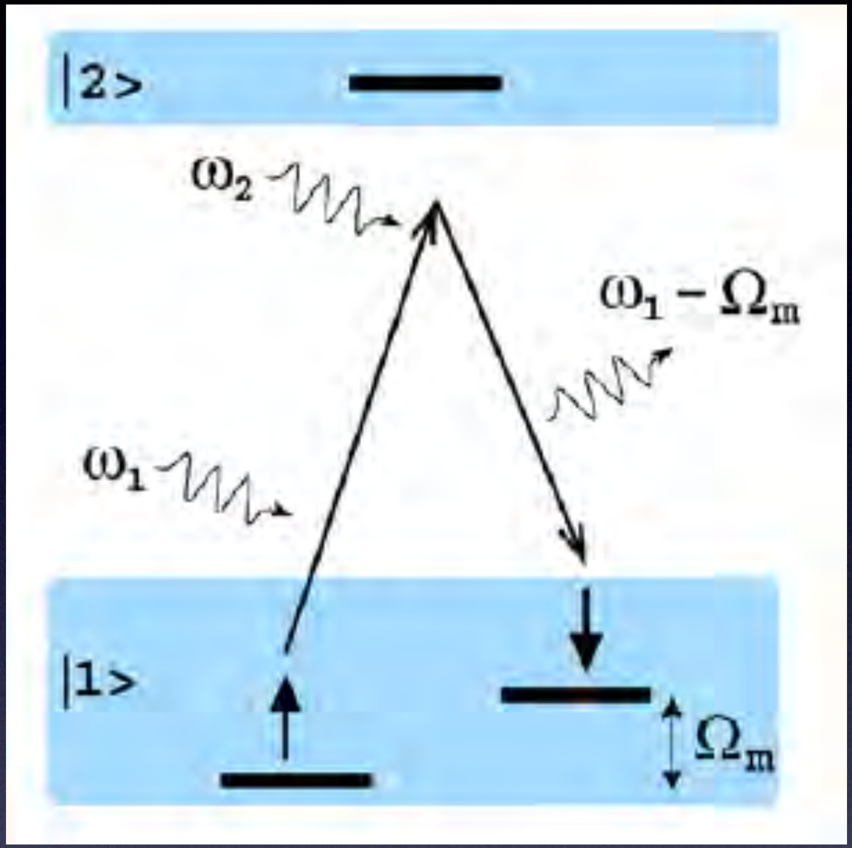


Zero-absorption regime of spin dynamics

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

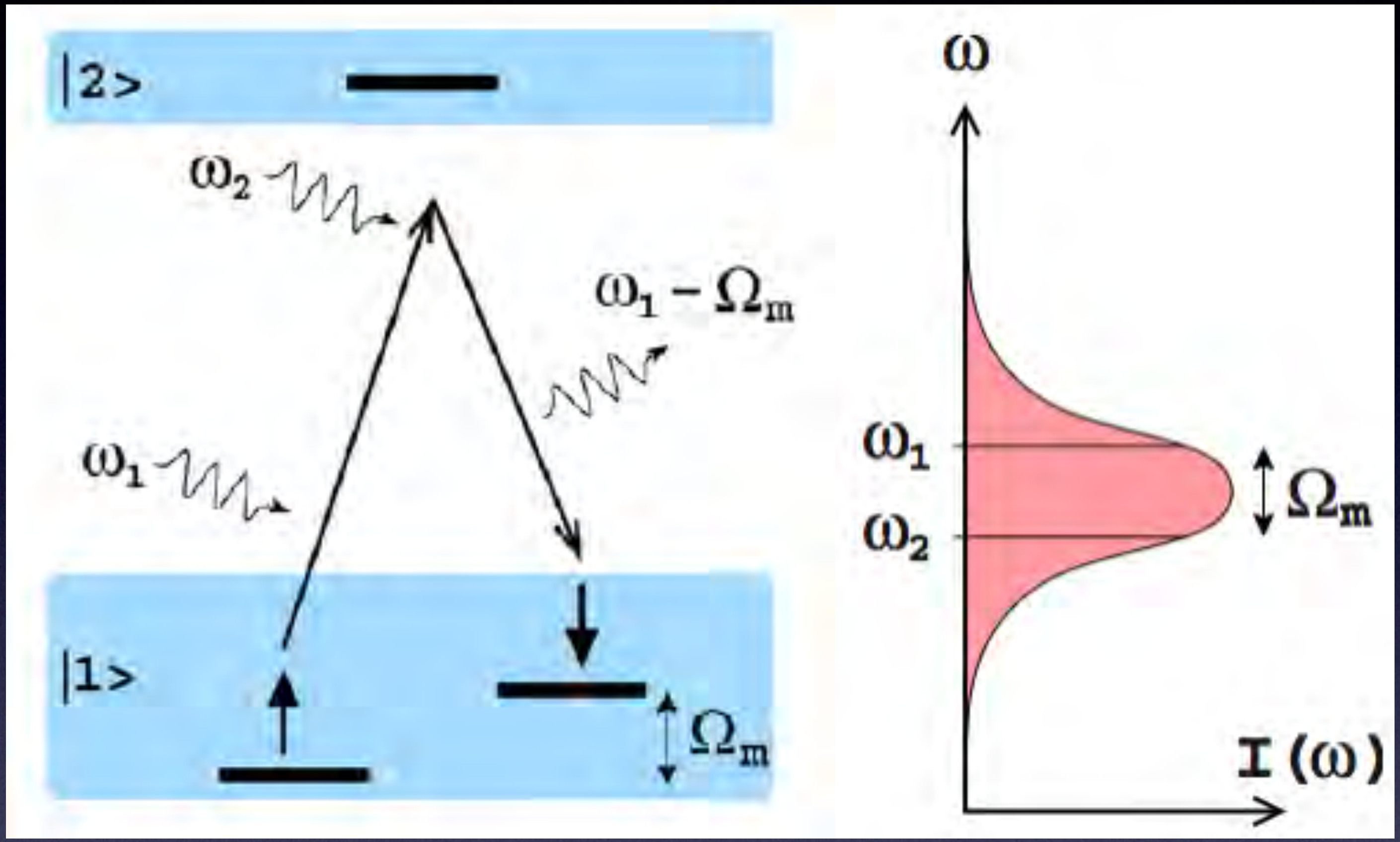
Impulsive Stimulated Raman

ISRS



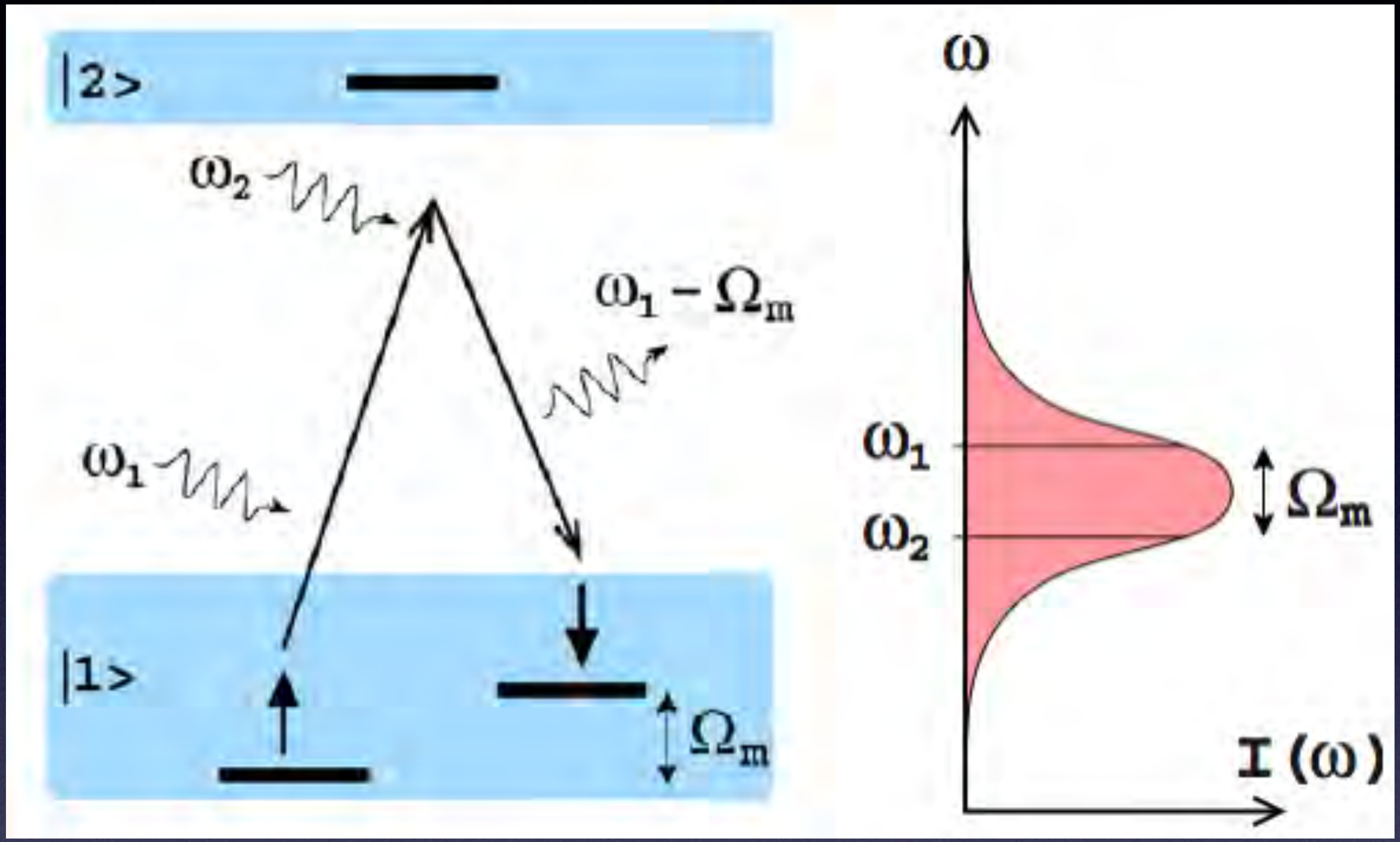
Impulsive Stimulated Raman

ISRS



Impulsive Stimulated Raman

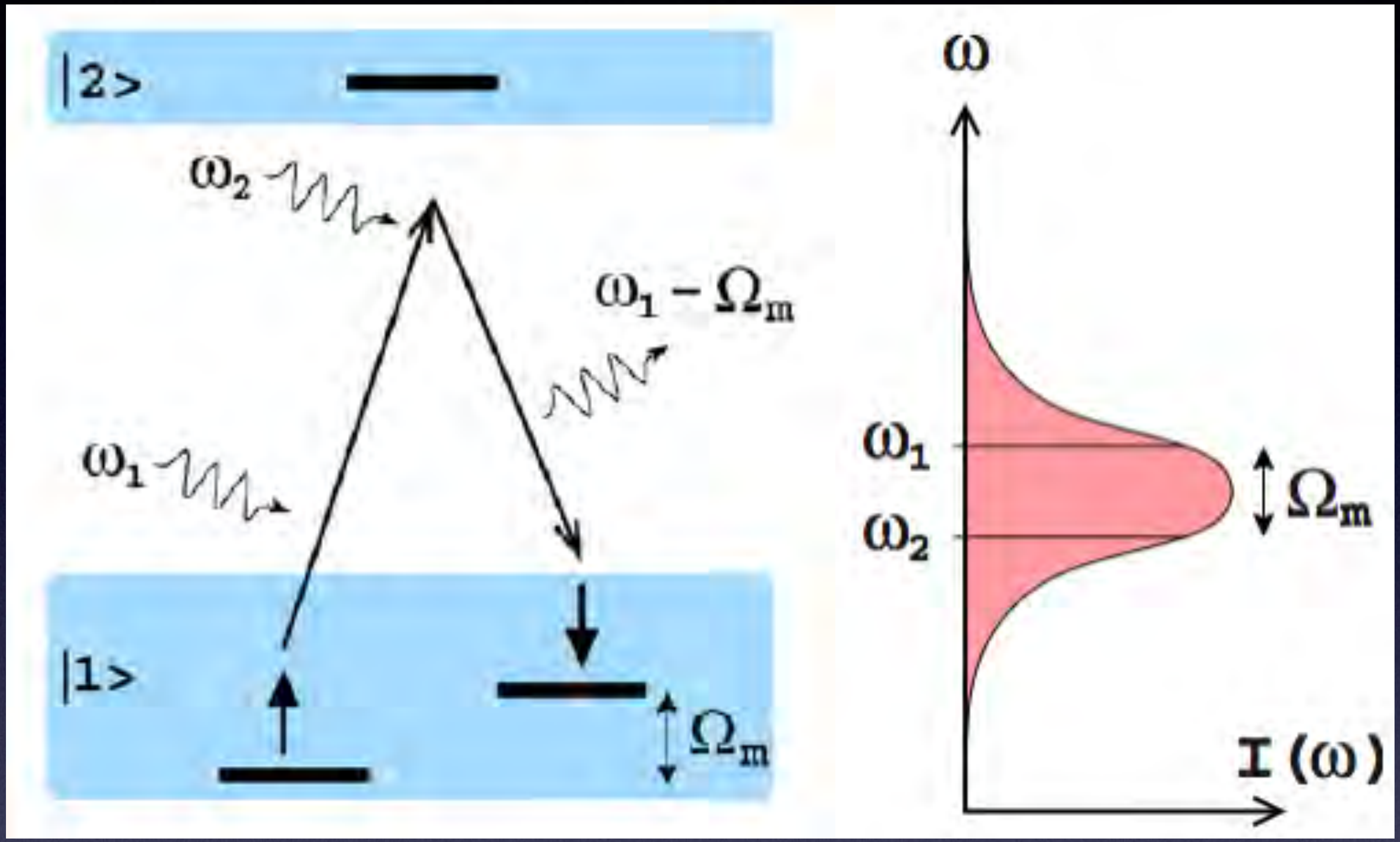
ISRS



Pulses shorter than period

Impulsive Stimulated Raman

ISRS



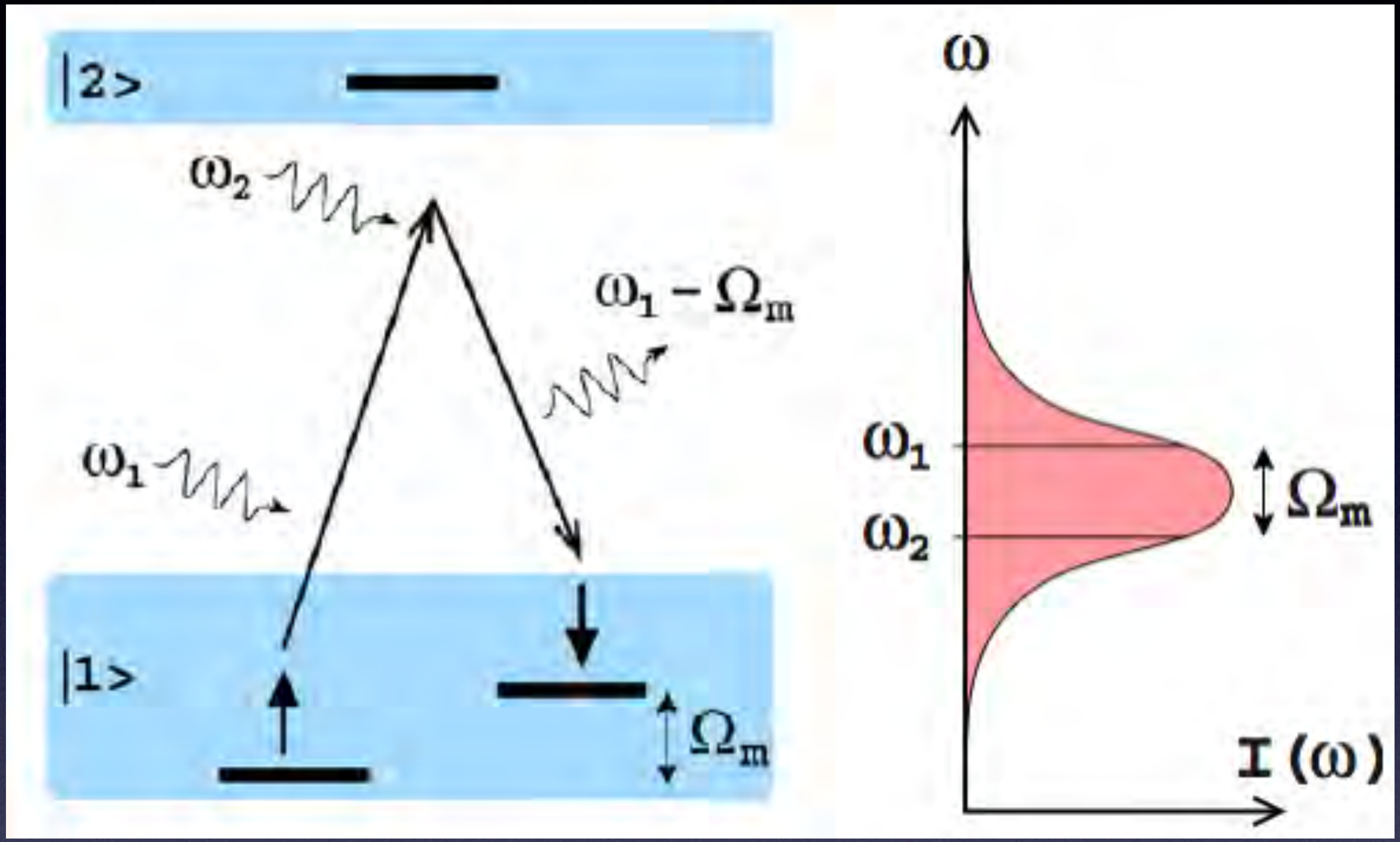
2M period in $KNiF_3$: 45 fs

Time domain

Pulses shorter than period

Impulsive Stimulated Raman

ISRS



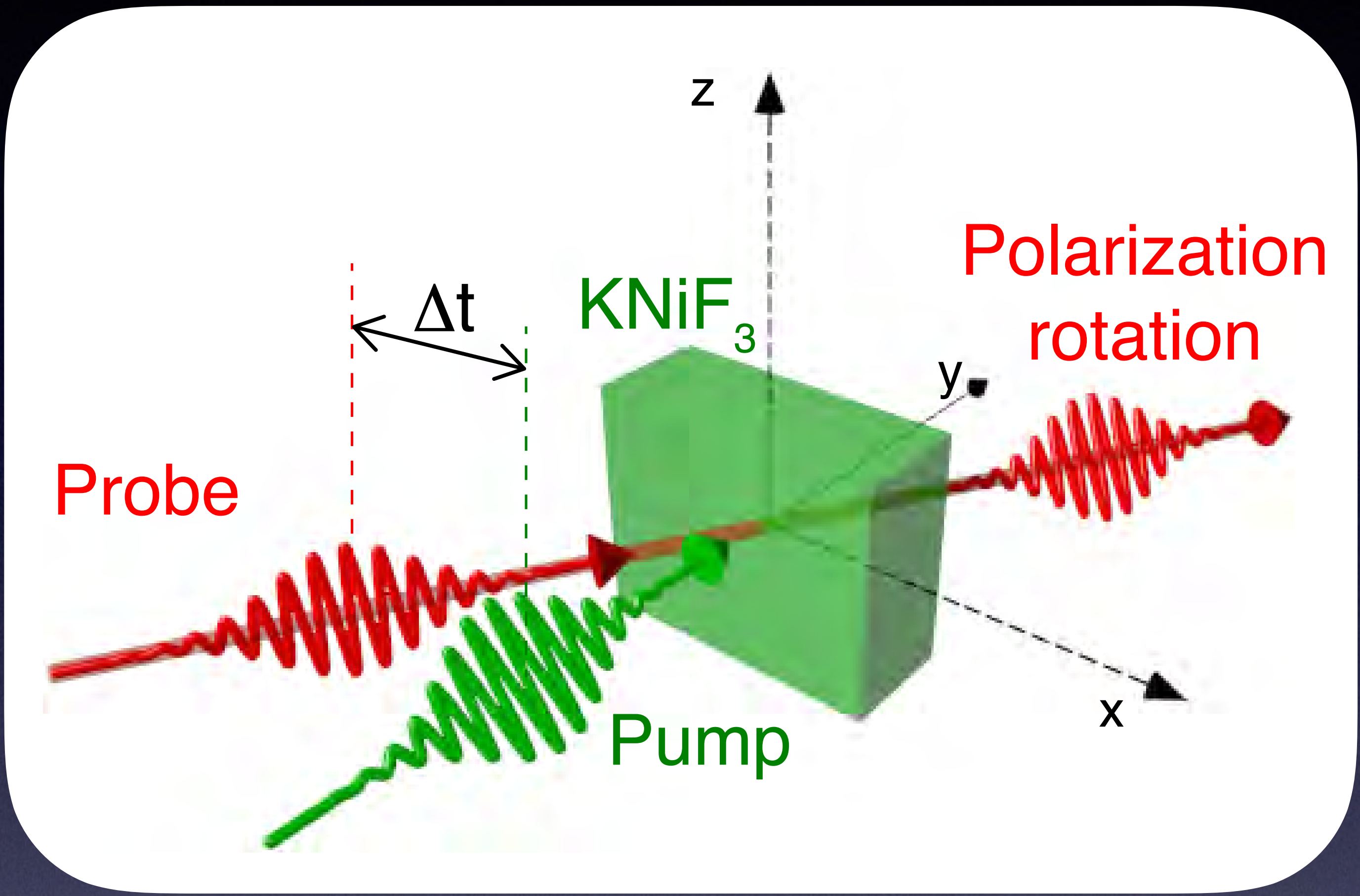
2M period in $KNiF_3$: 45 fs

Time domain

Pulses shorter than period

10 fs laser pulses

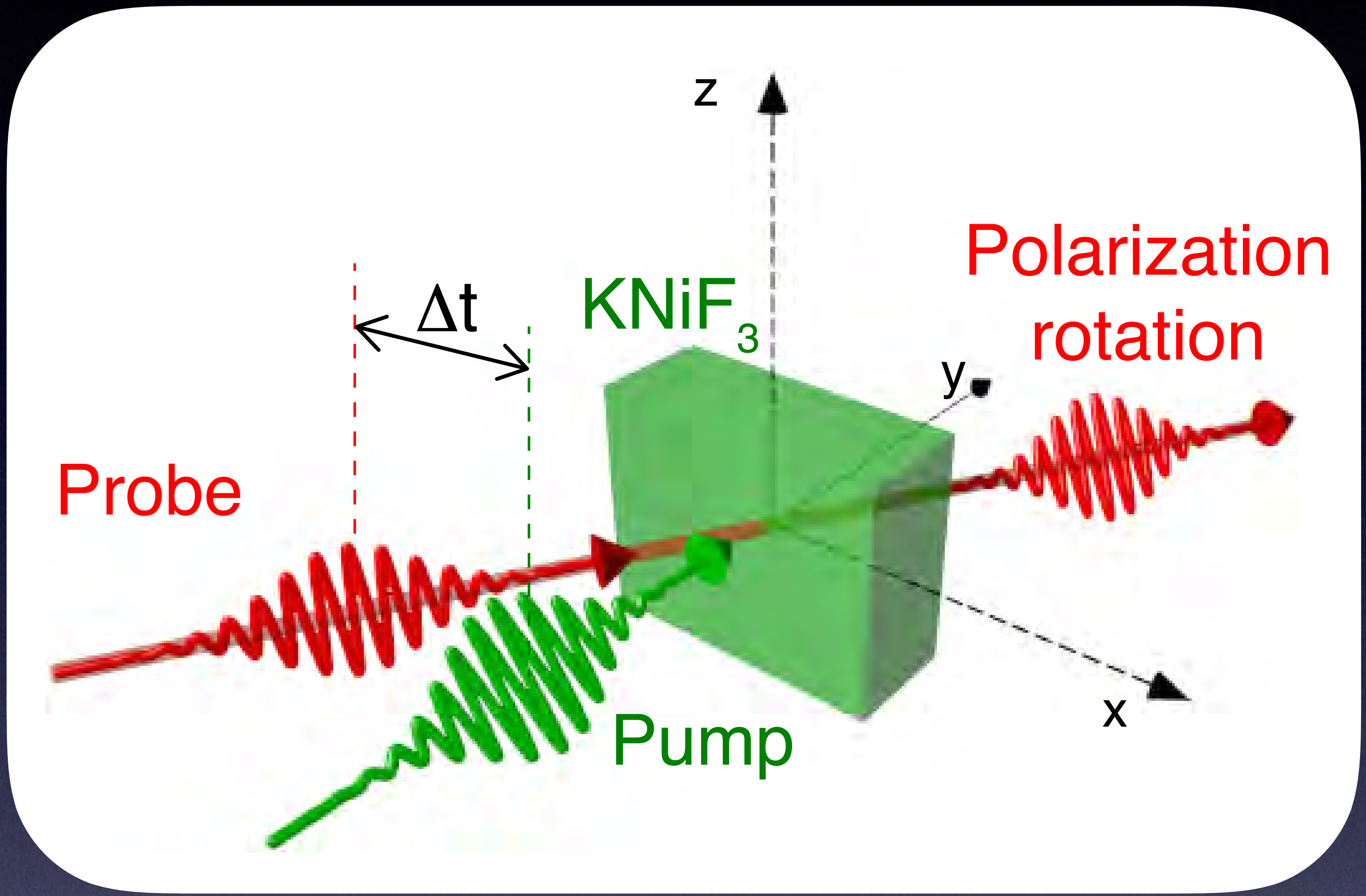
Detection



Pump-probe technique

Magneto-optical response to the photo-excitation measured as a function of the delay

Detection



Pump-probe technique

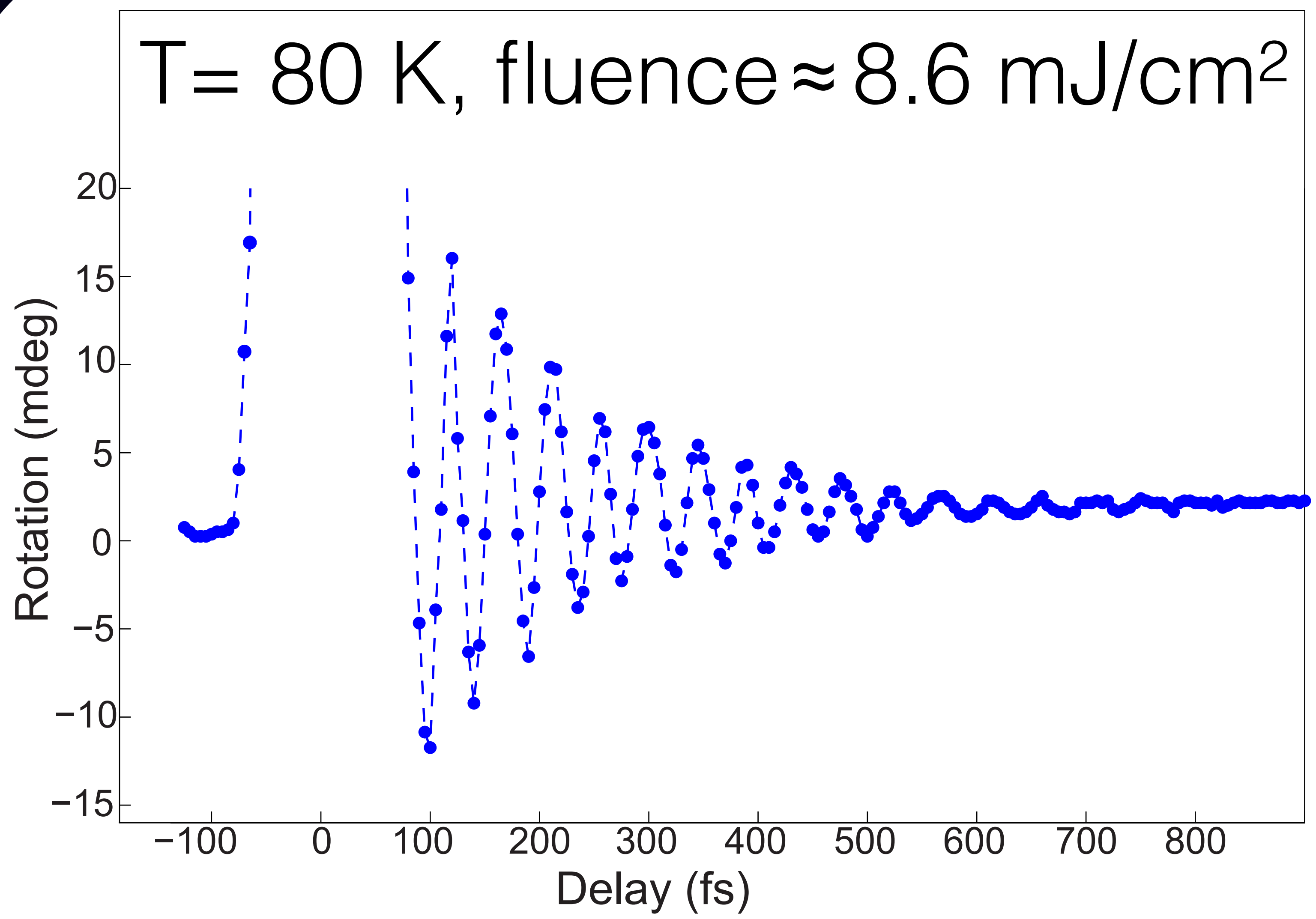
Magneto-optical response to the photo-excitation measured as a function of the delay

All-optical detection via a **second-order magneto-optical effect**

$$\epsilon_s^{\lambda\nu} = \sum_{ij} \sum_{\gamma\delta} \rho^{\lambda\nu\gamma\delta} \langle \hat{S}_i^{\gamma\Uparrow} \hat{S}_j^{\delta\Downarrow} \rangle$$

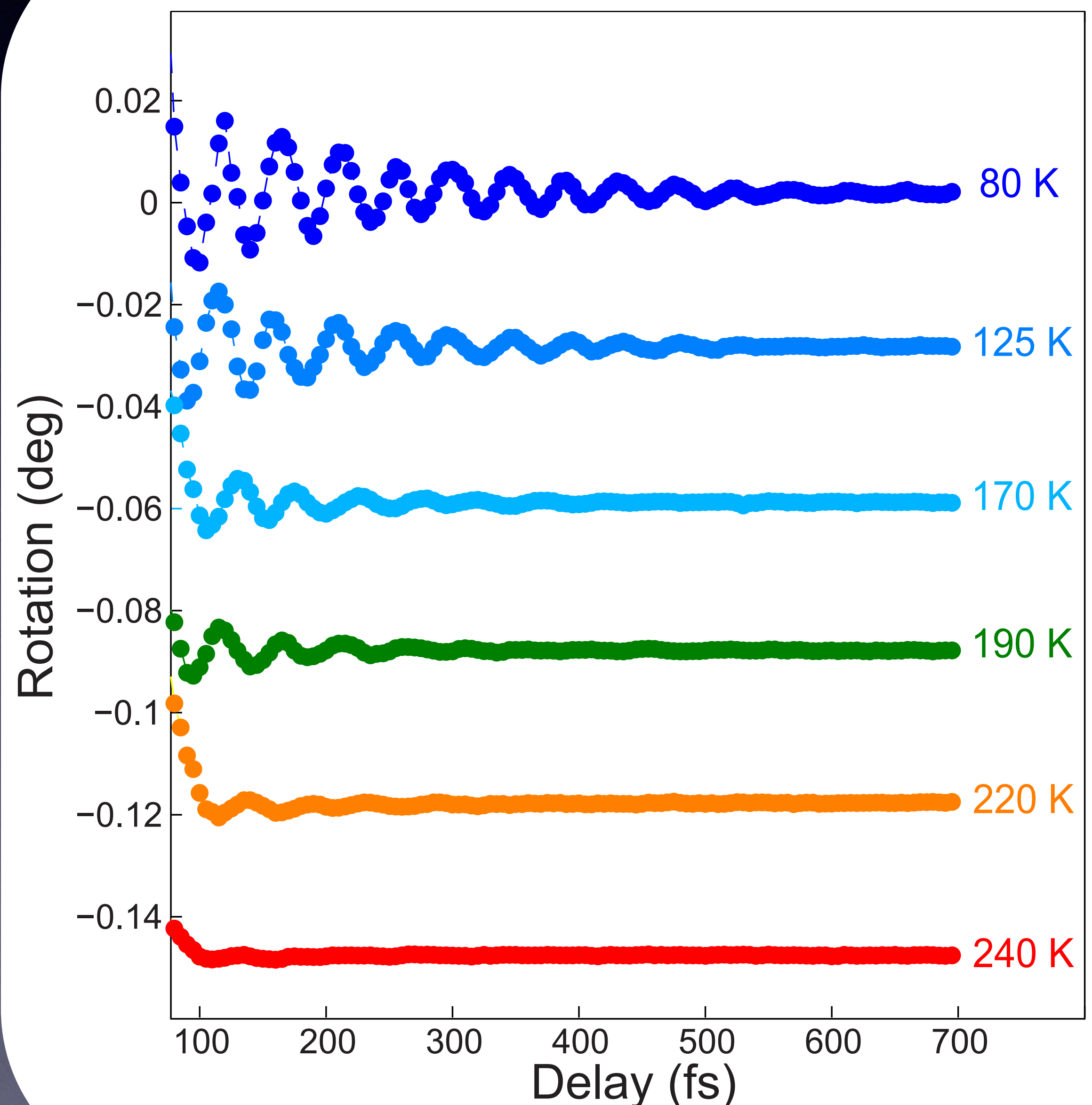
J. Ferrè *et al.* Rep. Prg. Phys **47**, 513 (1984)

Laser-induced dynamics

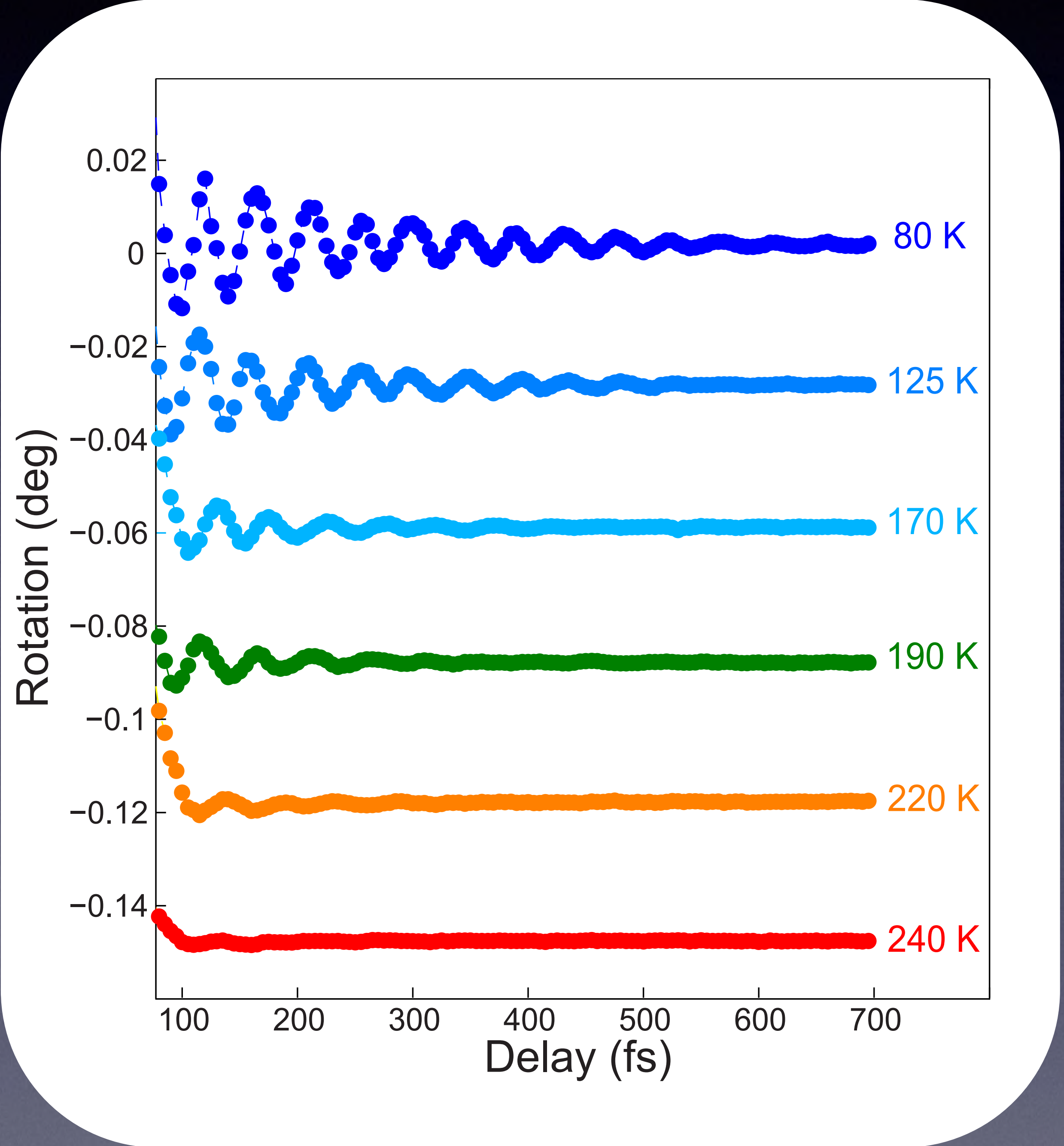


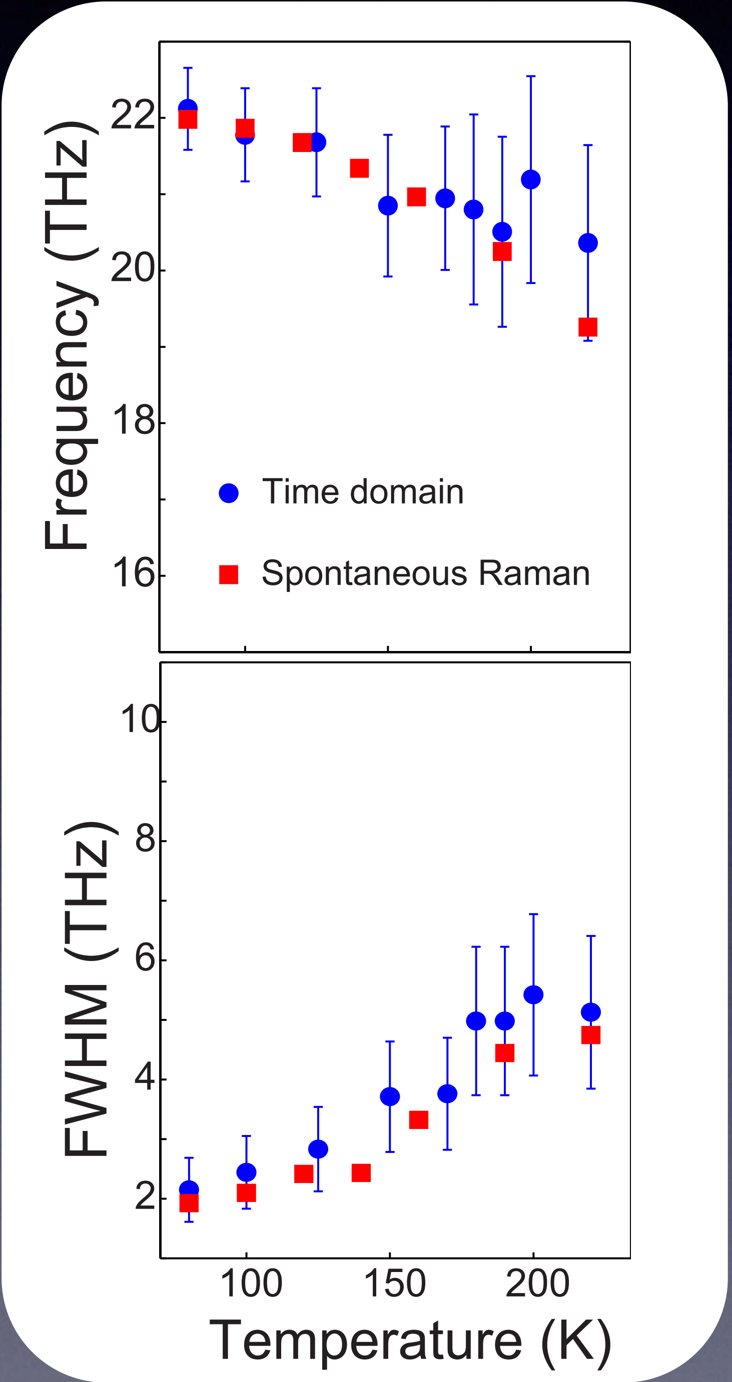
- ✓ Pump and probe linearly and orthogonally polarized
- ✓ Oscillations @ 22 THz (T=45 fs)
- ✓ Lifetime ≈ 500 fs

Temperature dependence

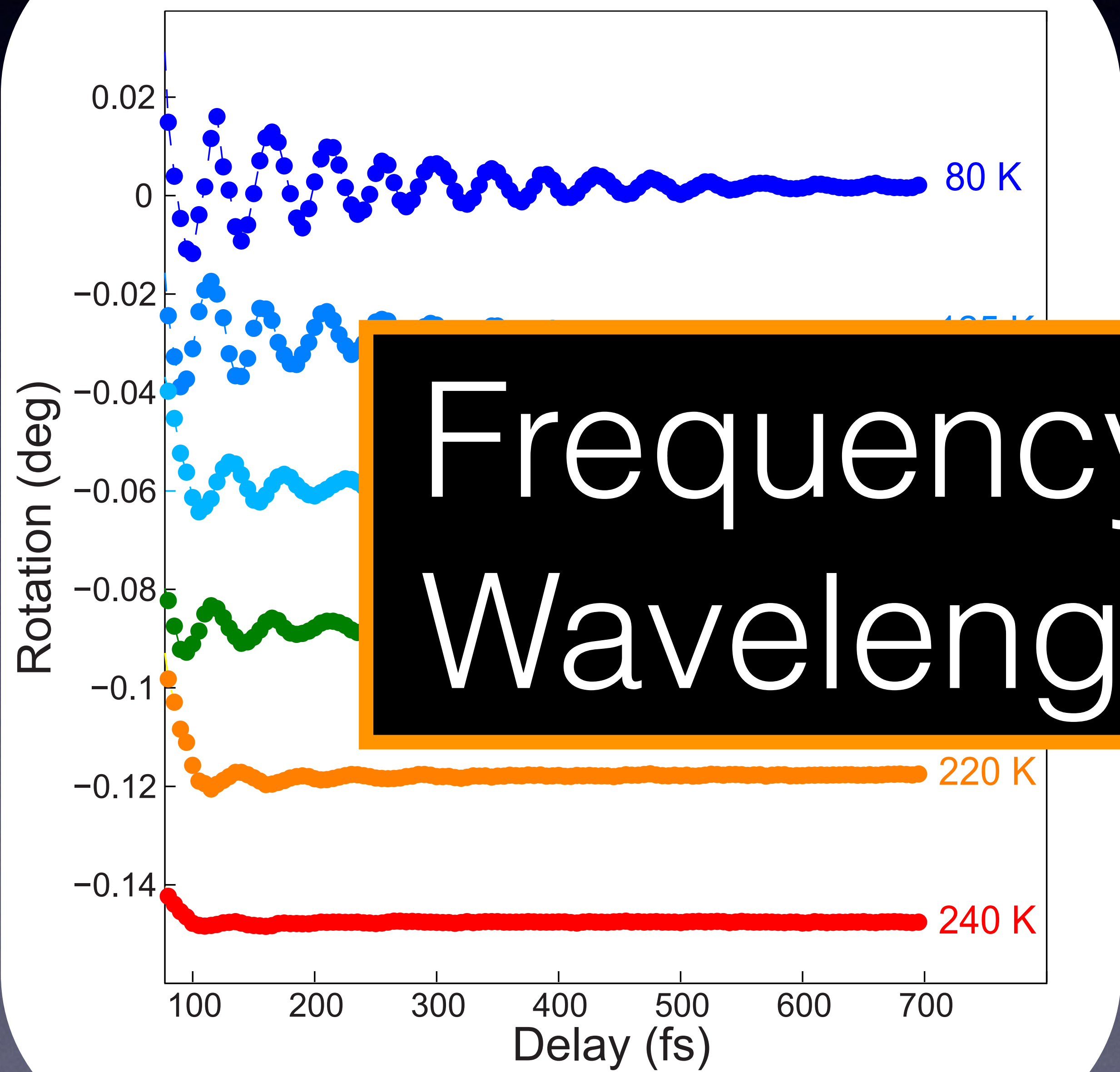


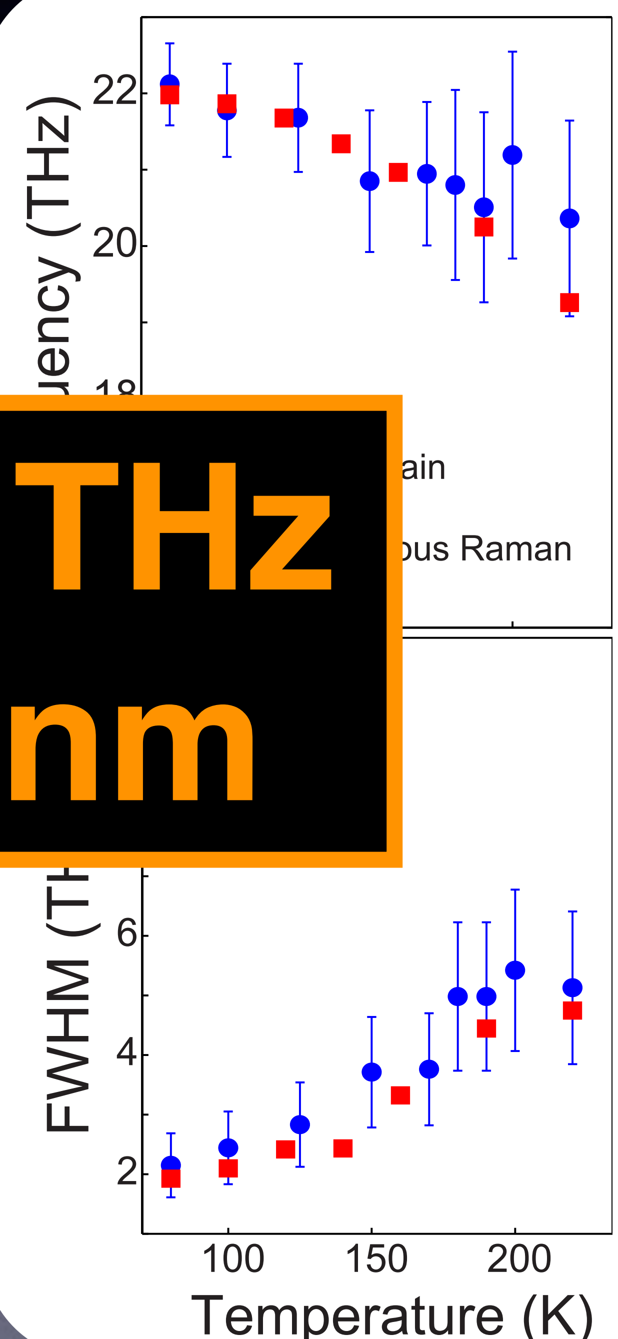
Temperature dependence





Temperature dependence





Frequency: **22 THz**
Wavelength: **1 nm**

Macrospin dynamics

$$\langle \hat{S}_i^{z\Uparrow} \hat{S}_j^{z\Downarrow} \rangle \qquad L^z(t)$$

Macrospin dynamics

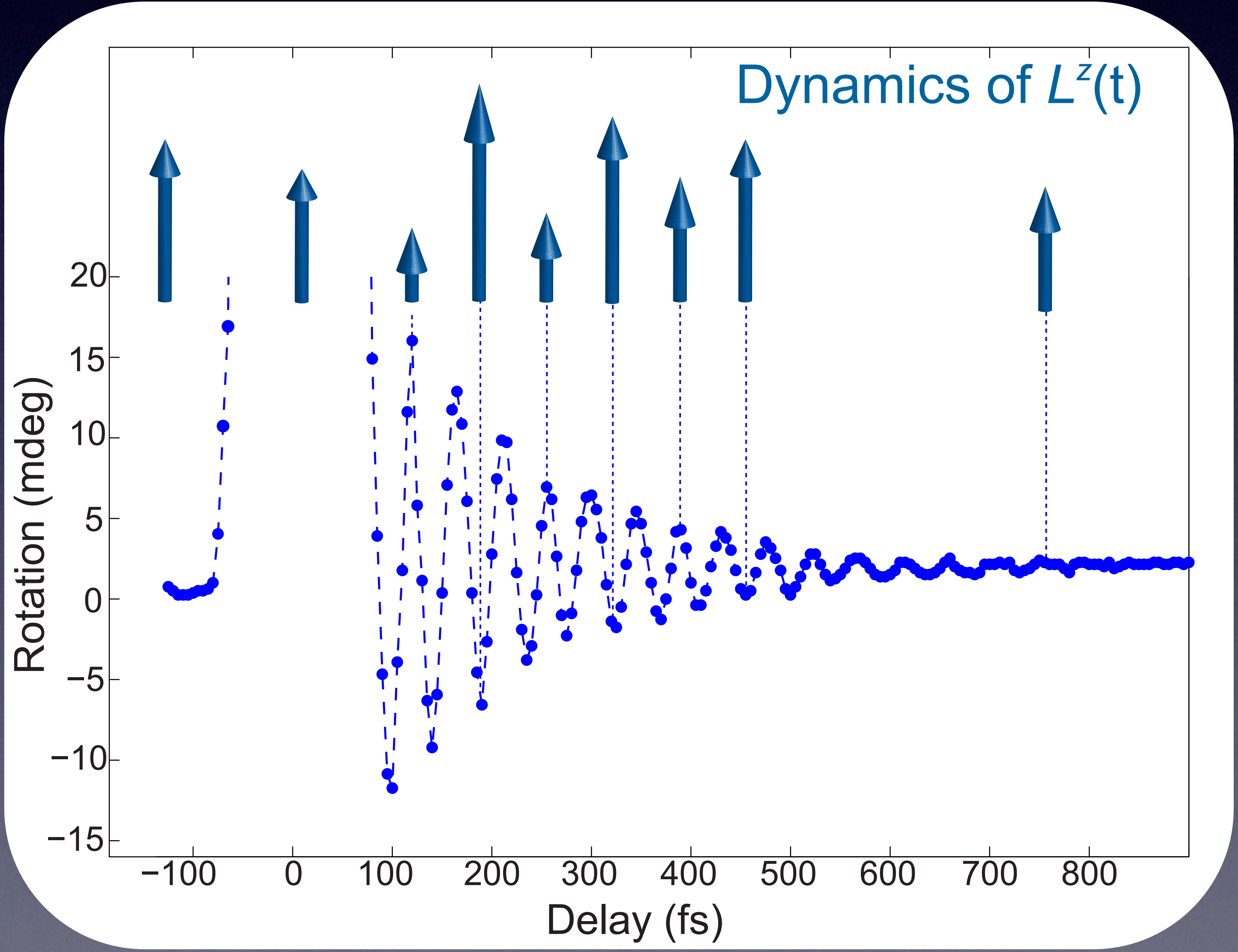
$$\langle \hat{S}_i^{z\Uparrow} \hat{S}_j^{z\Downarrow} \rangle \qquad L^z(t)$$

Same time-dependence

Macrospin dynamics

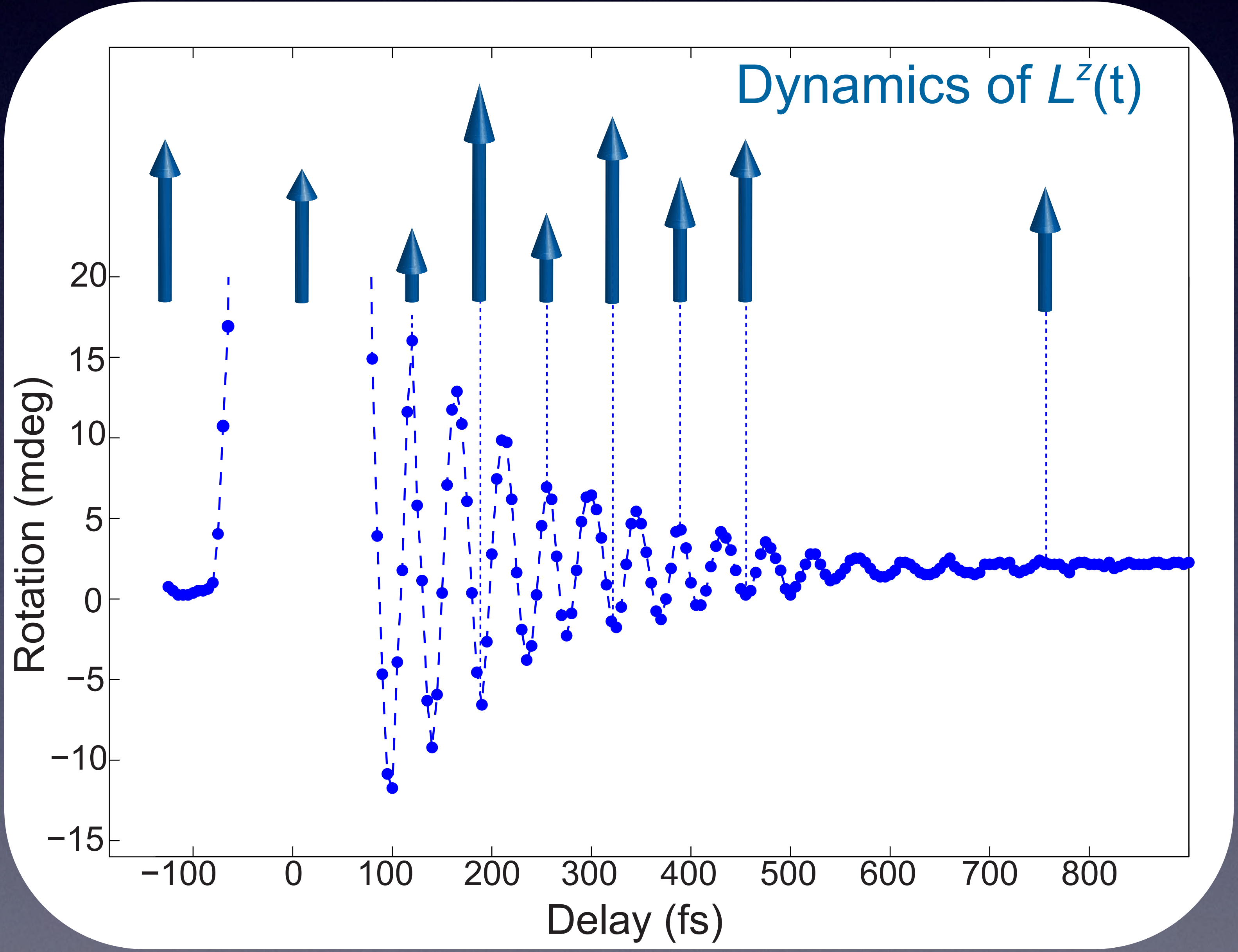
$\langle \hat{S}_i^{z\Uparrow} \hat{S}_j^{z\Downarrow} \rangle \qquad L^z(t)$

Same time-dependence



Macrospin dynamics

$$\langle \hat{S}_i^{z\Uparrow} \hat{S}_j^{z\Downarrow} \rangle \qquad L^z(t)$$

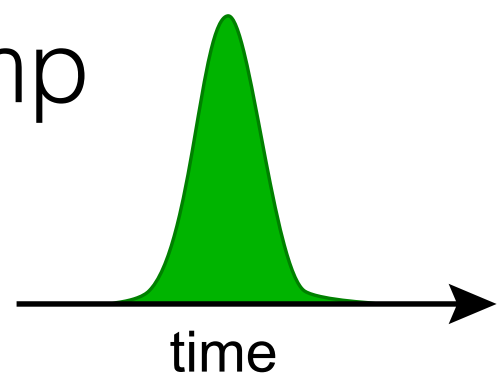


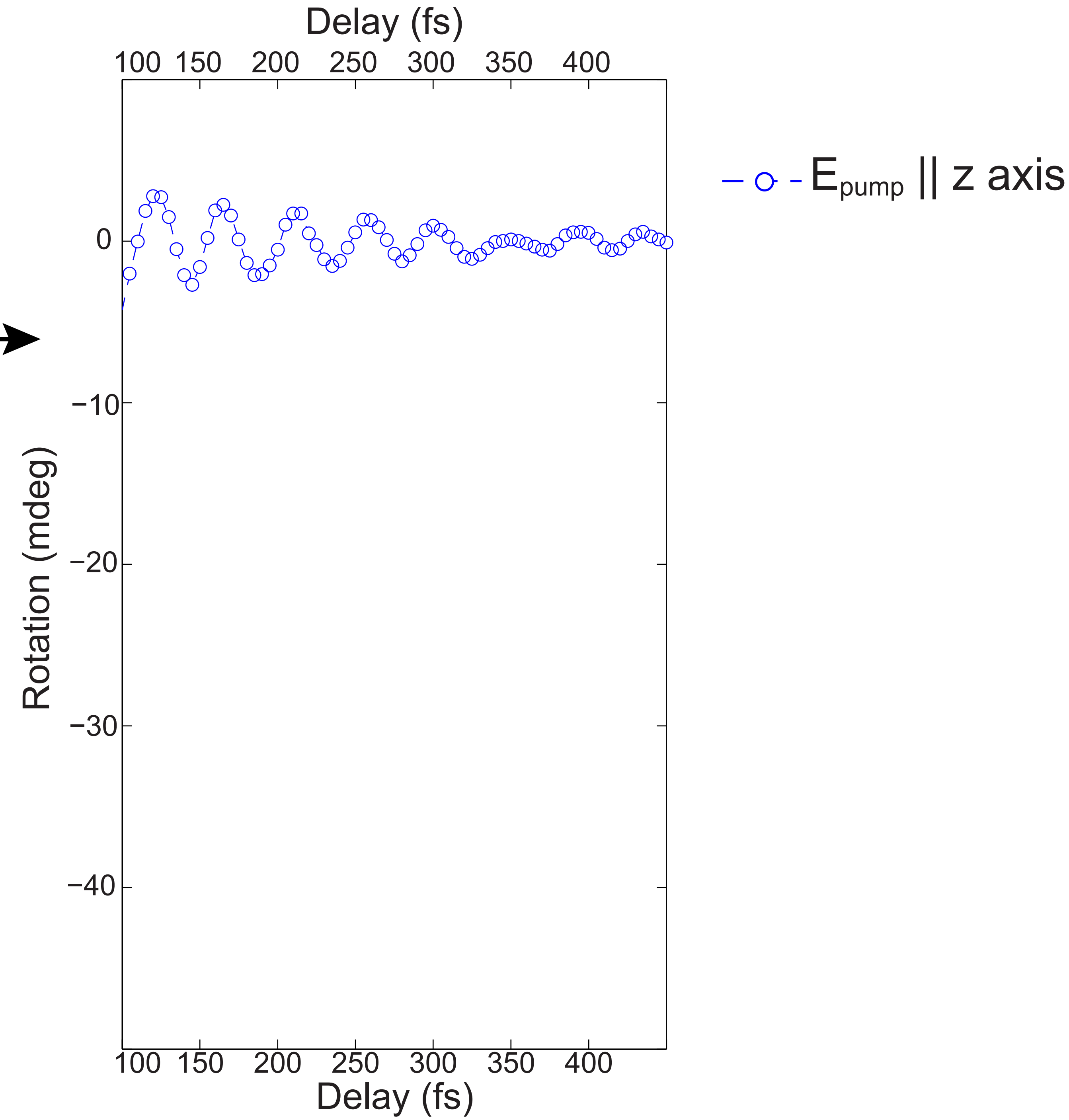
Same time-dependence

Macroscopic probe of the femtosecond dynamics of nanometer spin correlations

Control the dynamics

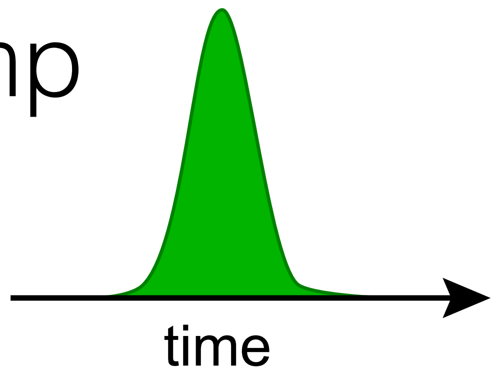
One pump pulse

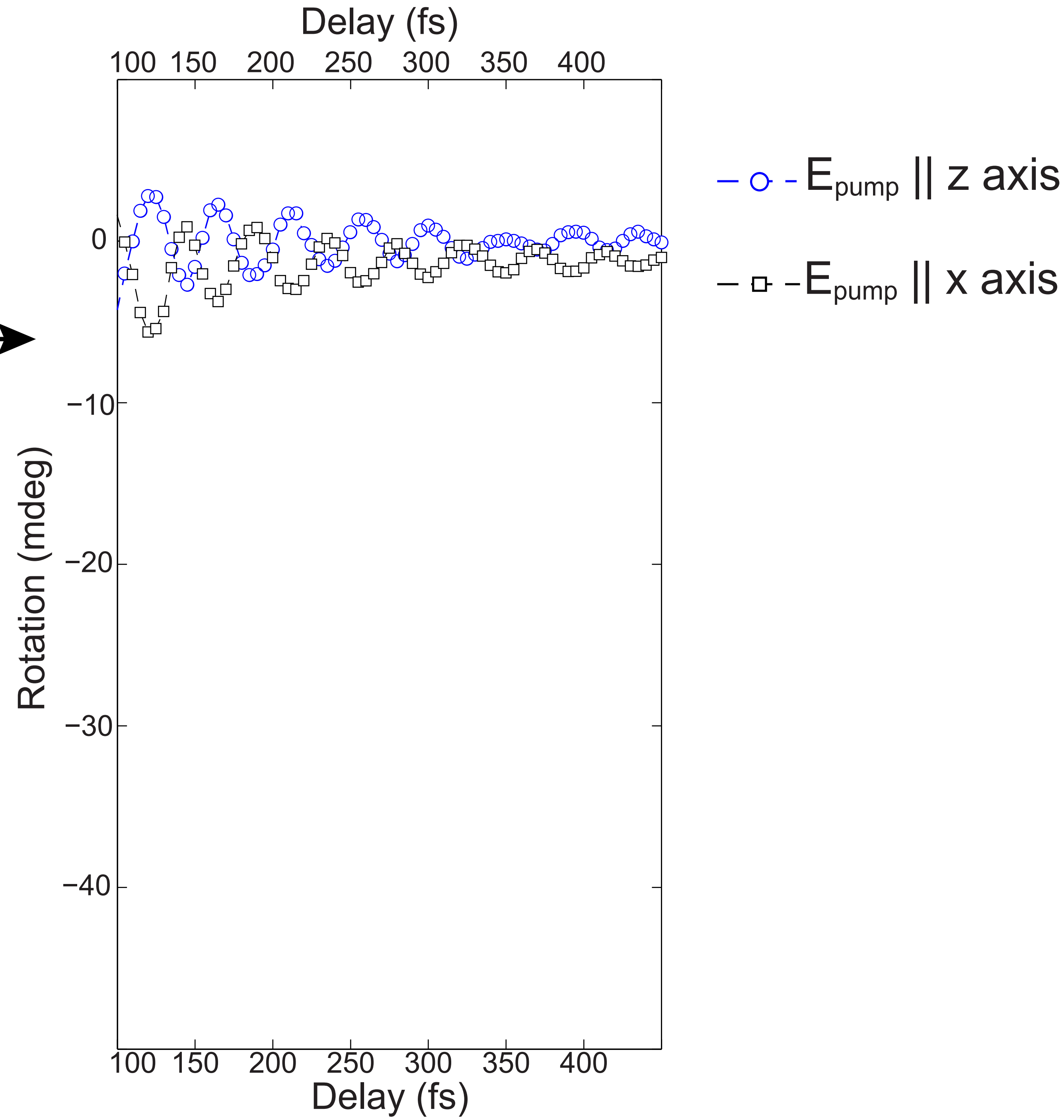




Control the dynamics

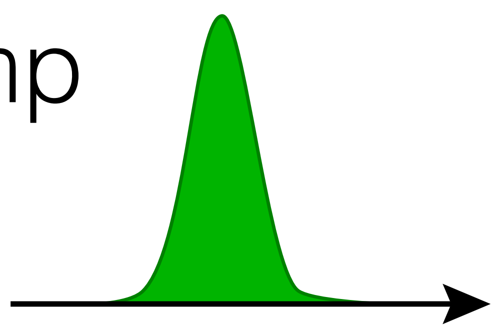
One pump pulse

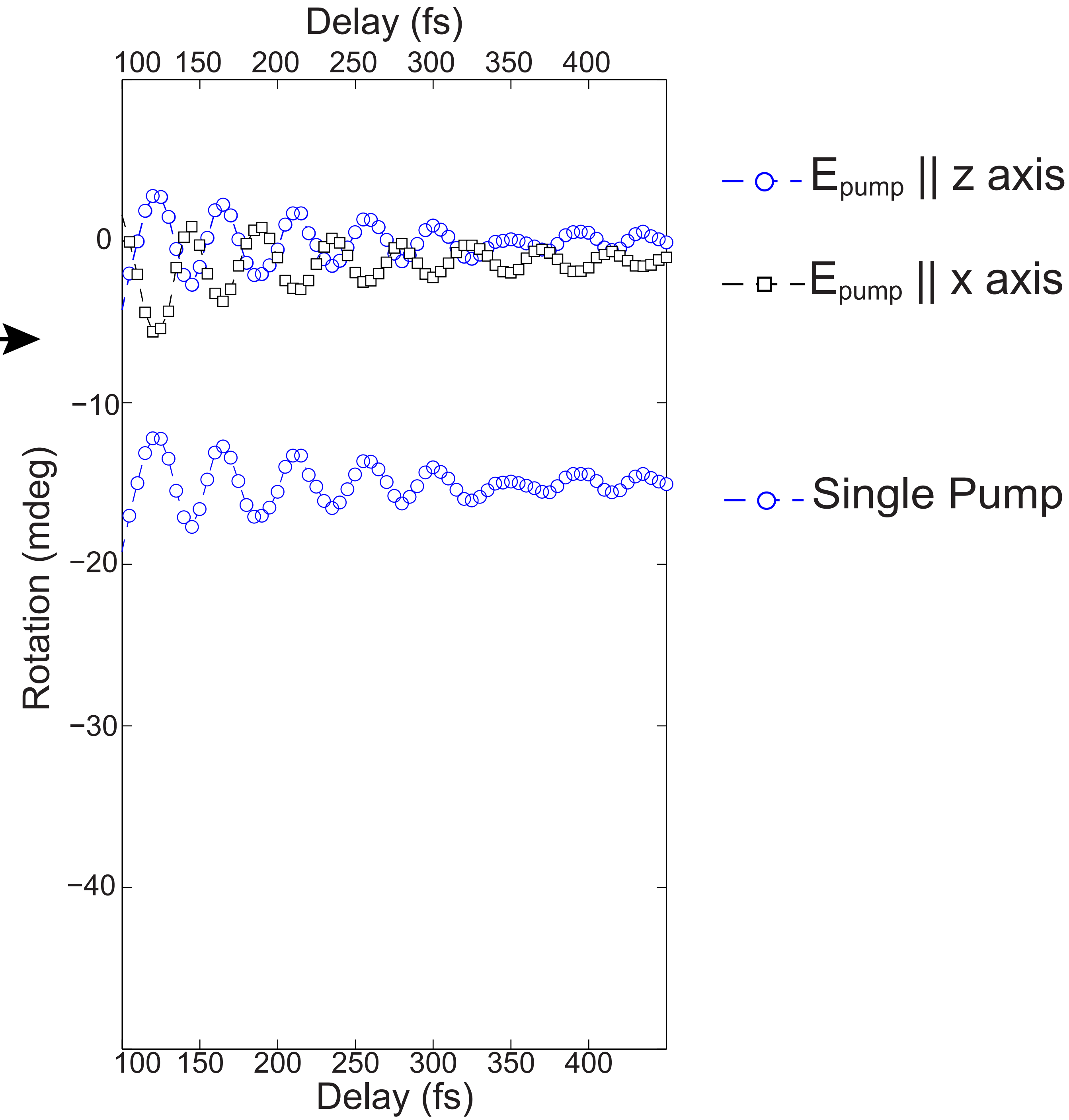




Control the dynamics

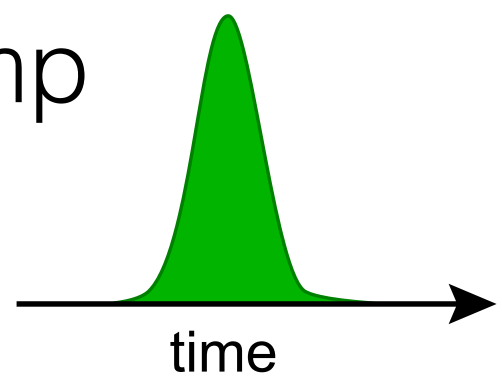
One pump pulse



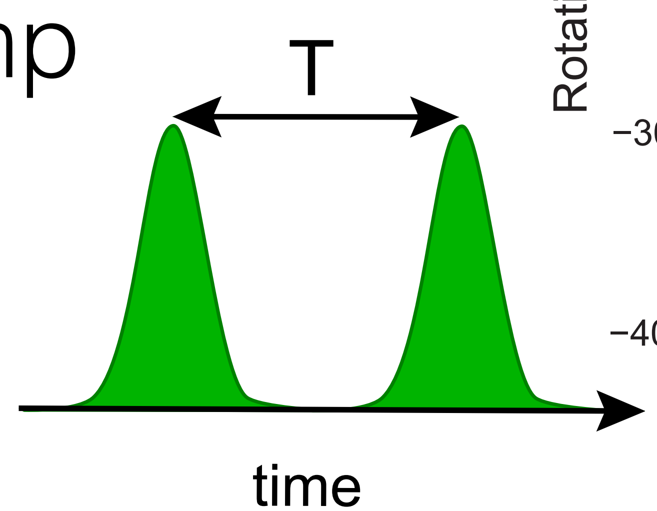


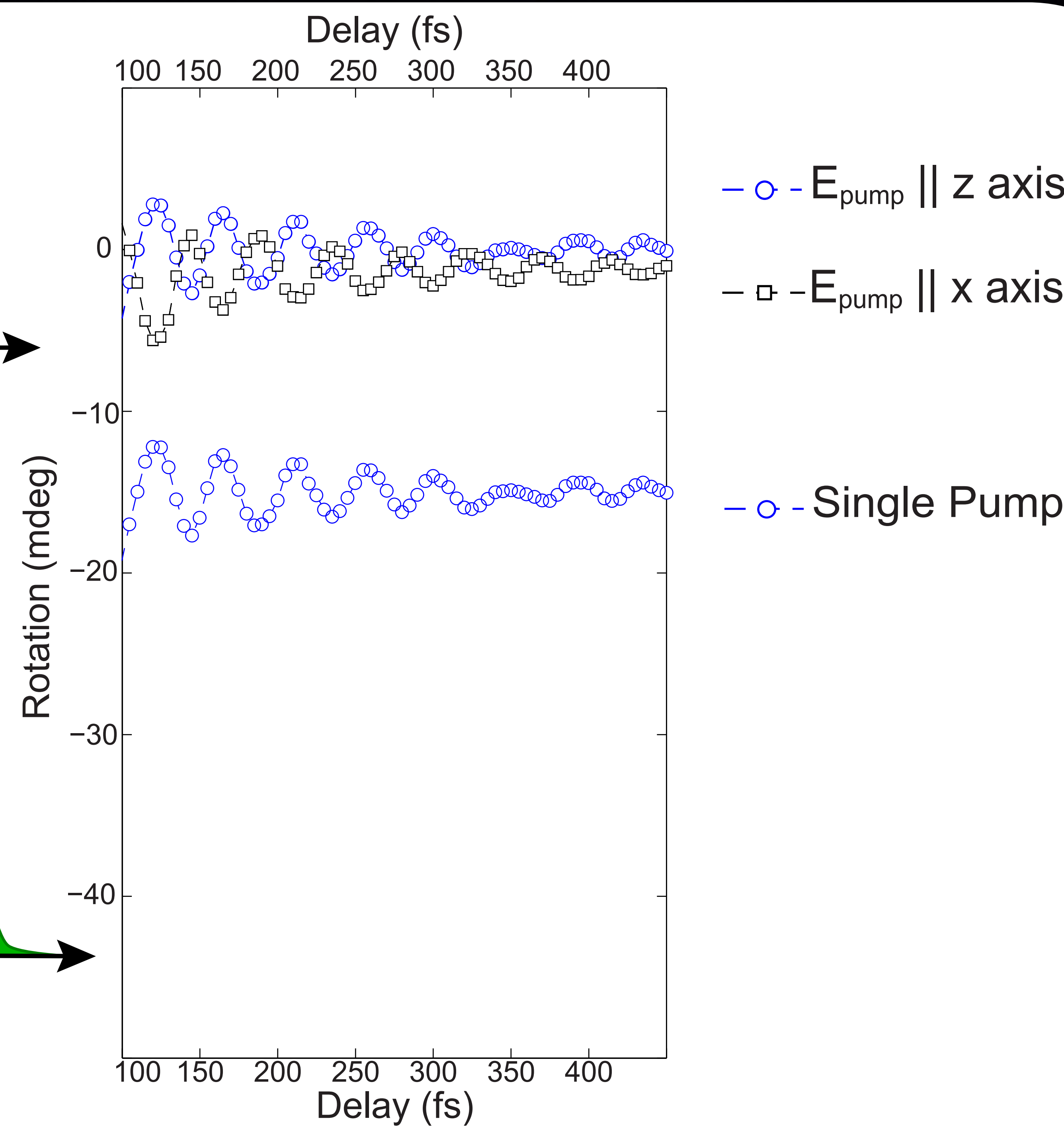
Control the dynamics

One pump pulse

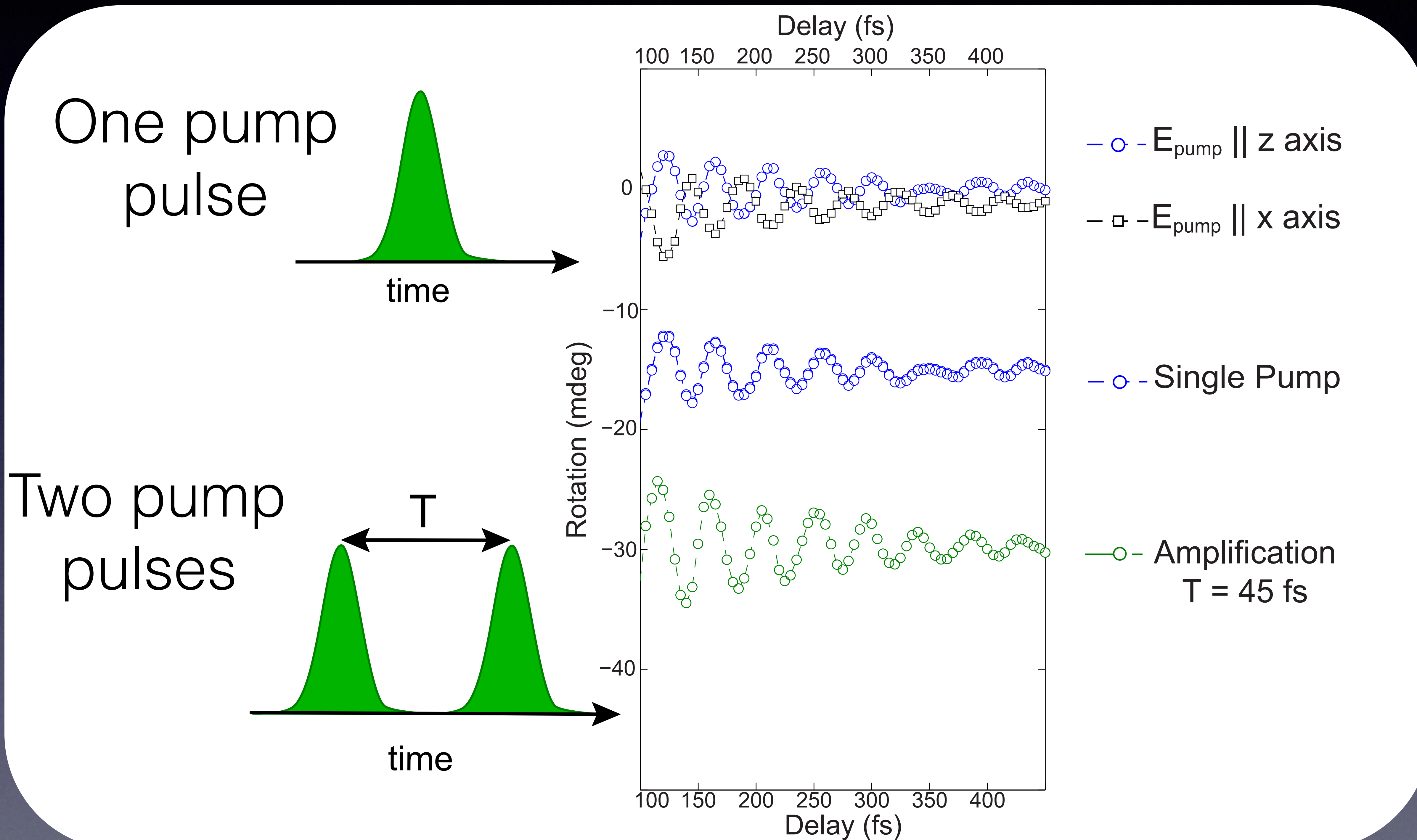


Two pump pulses

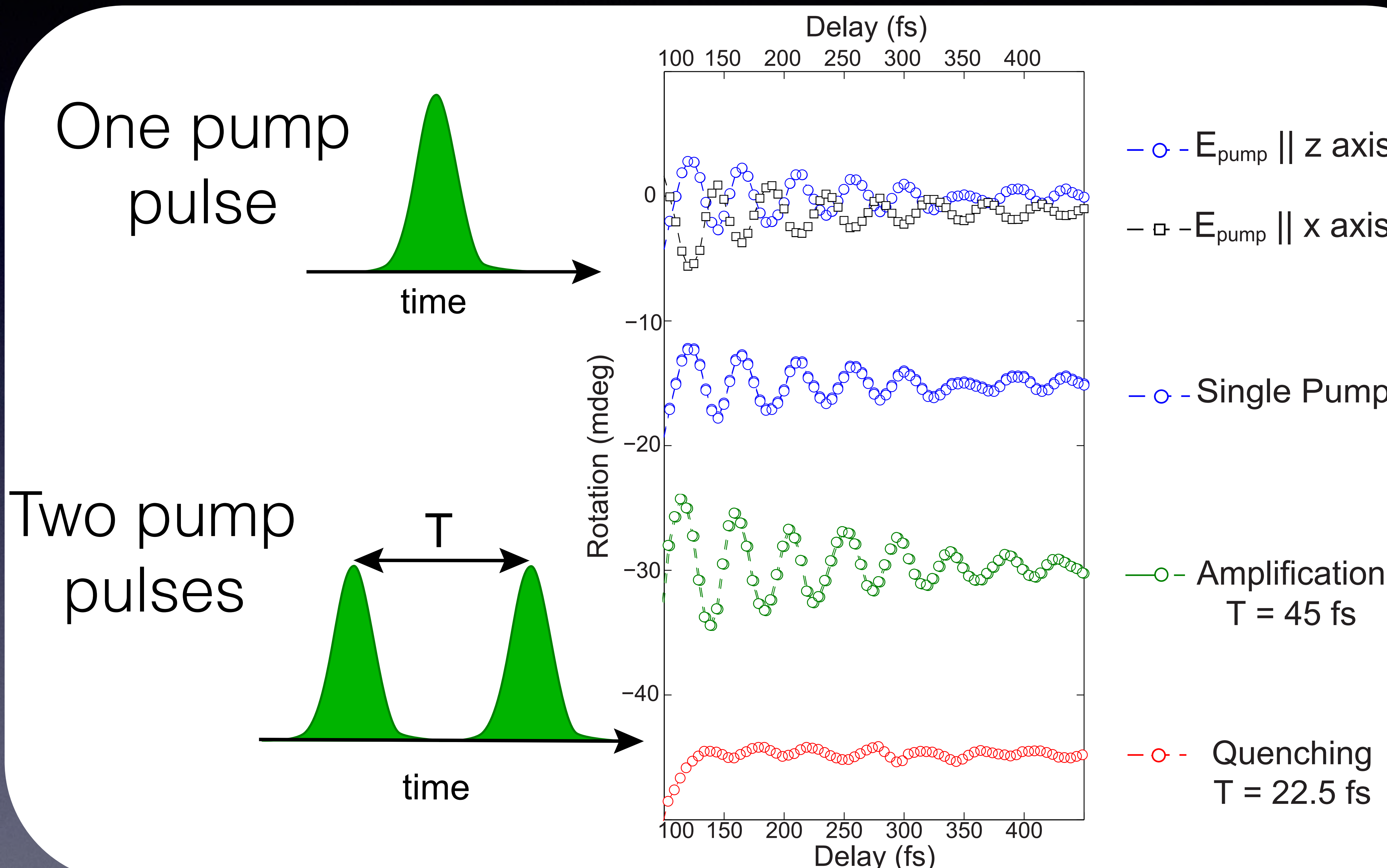




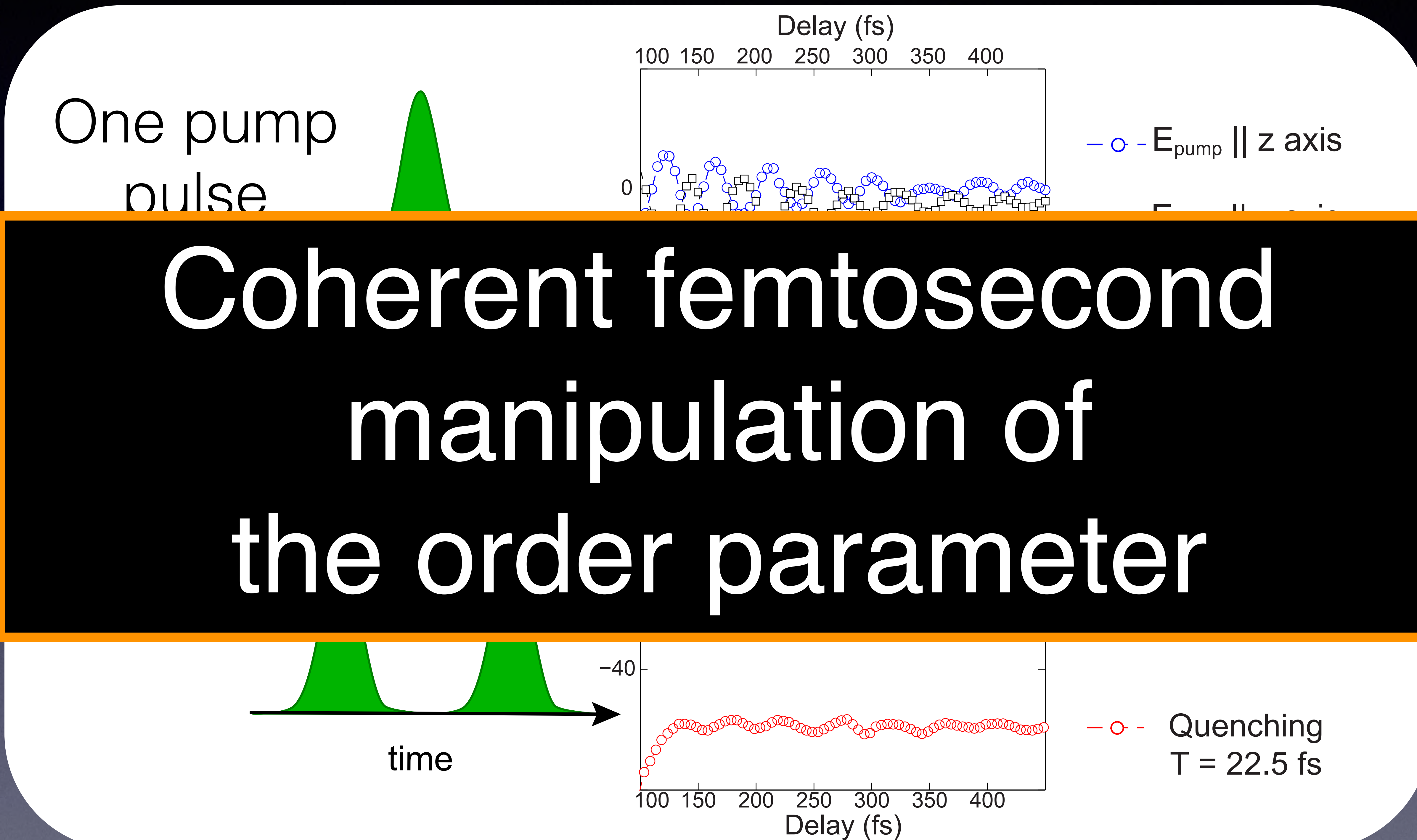
Control the dynamics



Control the dynamics

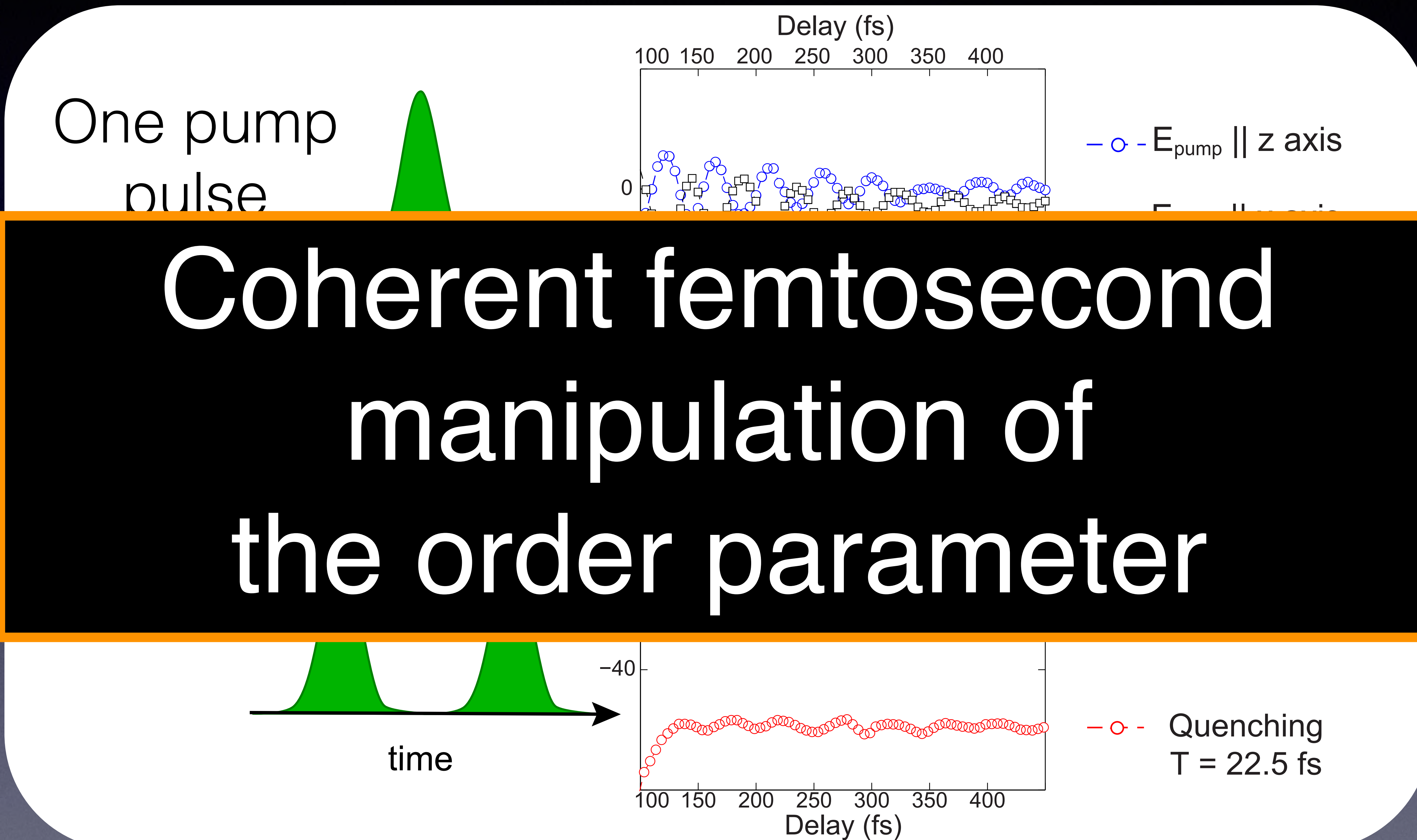


Control the dynamics



Coherent femtosecond manipulation of the order parameter

Control the dynamics



Coherent femtosecond manipulation of the order parameter

D. Bossini et al. Nat. Comm. **7,** *10645 (2016)*

Length-scale dynamics

Nearest-neighbors correlations

Length-scale dynamics

Nearest-neighbors correlations

Counterintuitive: MO macroscopic probe

Length-scale dynamics

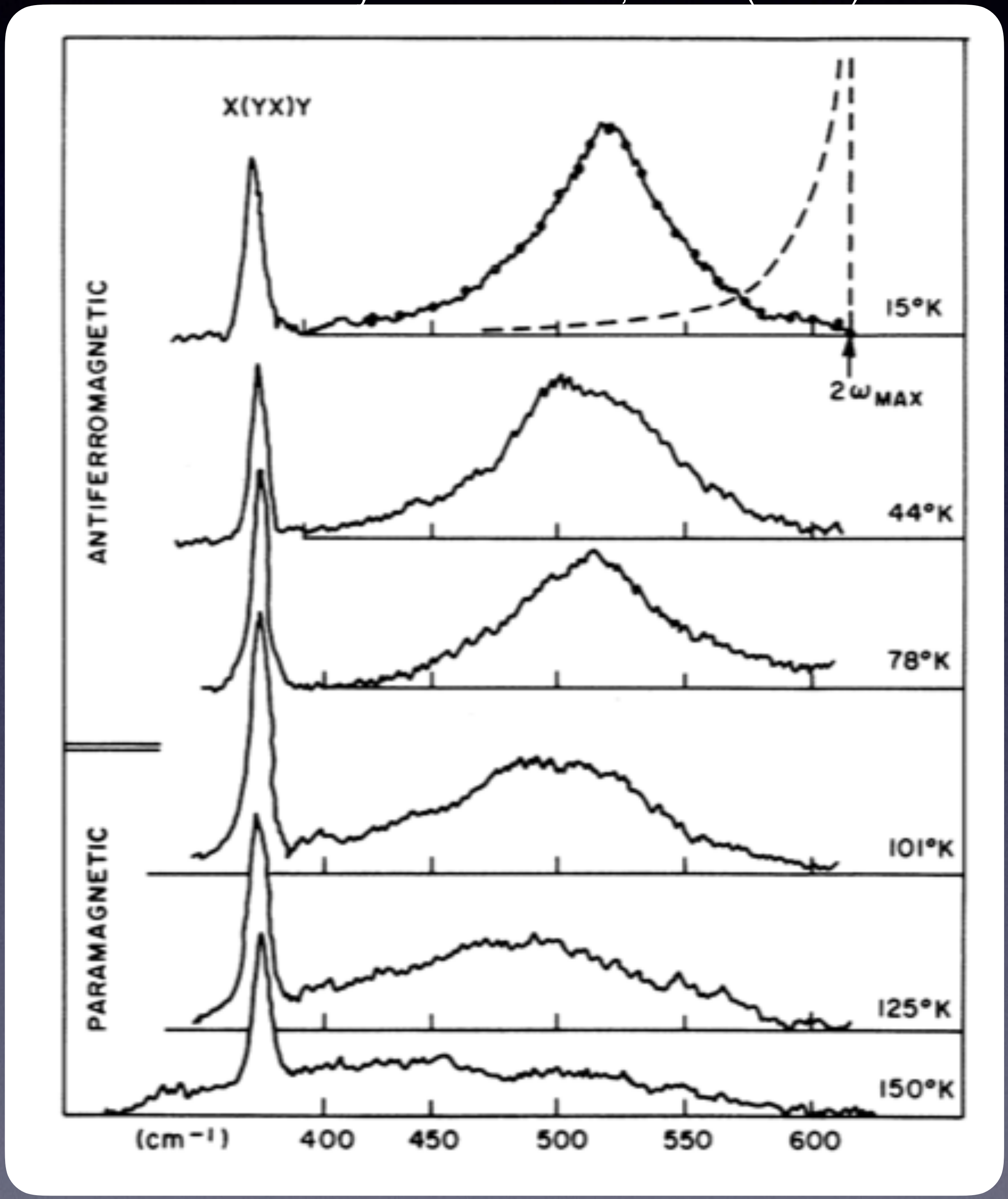
Nearest-neighbors correlations

Counterintuitive: MO macroscopic probe

Experimental evidence of
short-range nature
of the interaction ?

Length-scale dynamics

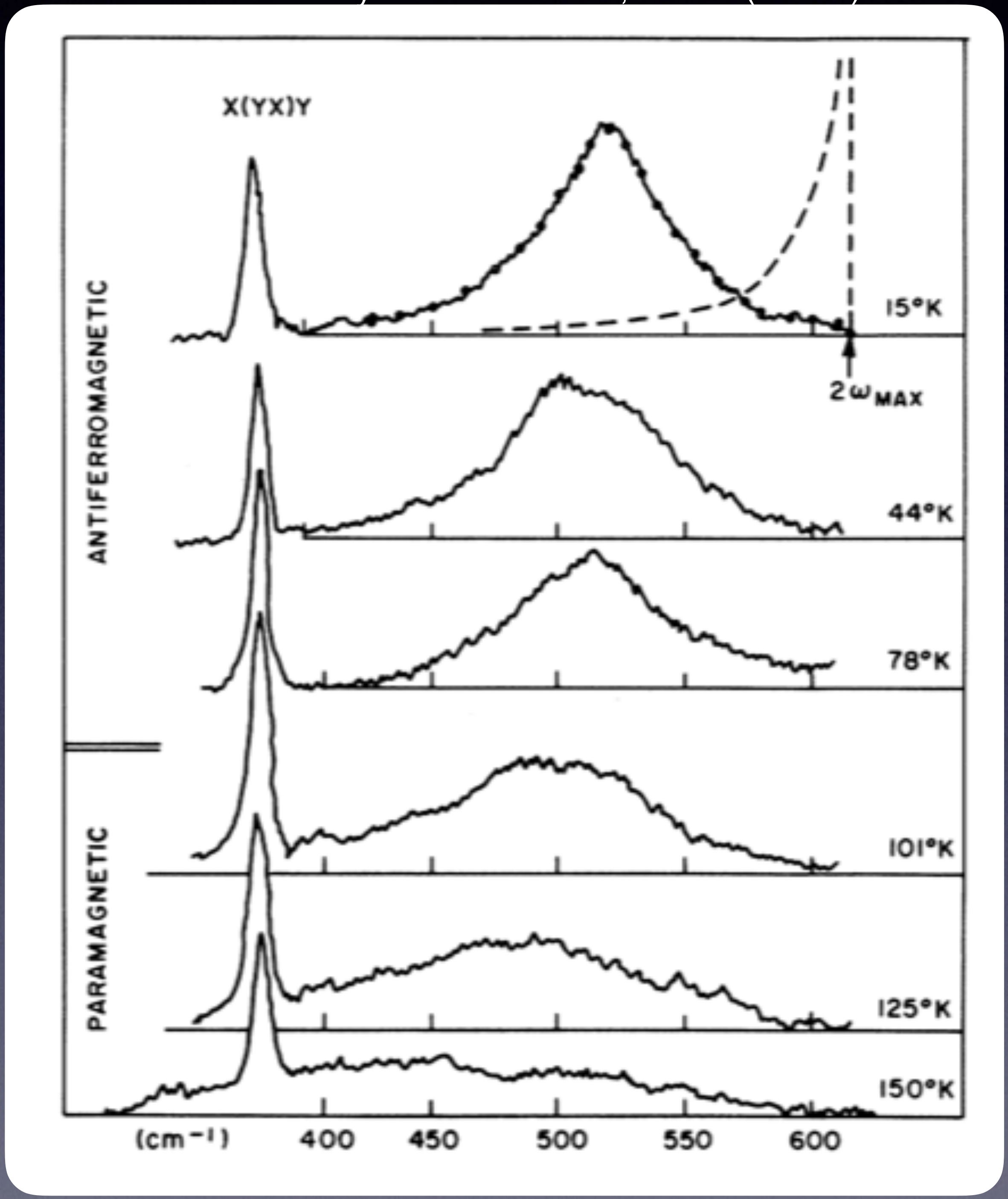
P. Fleury *et al.* PRL **24**, 1347 (1970)



- K_2NiF_4, T_N = 96 K
- 2M-mode up to T ~ 1.5 x T_N

Length-scale dynamics

P. Fleury *et al.* PRL **24**, 1347 (1970)

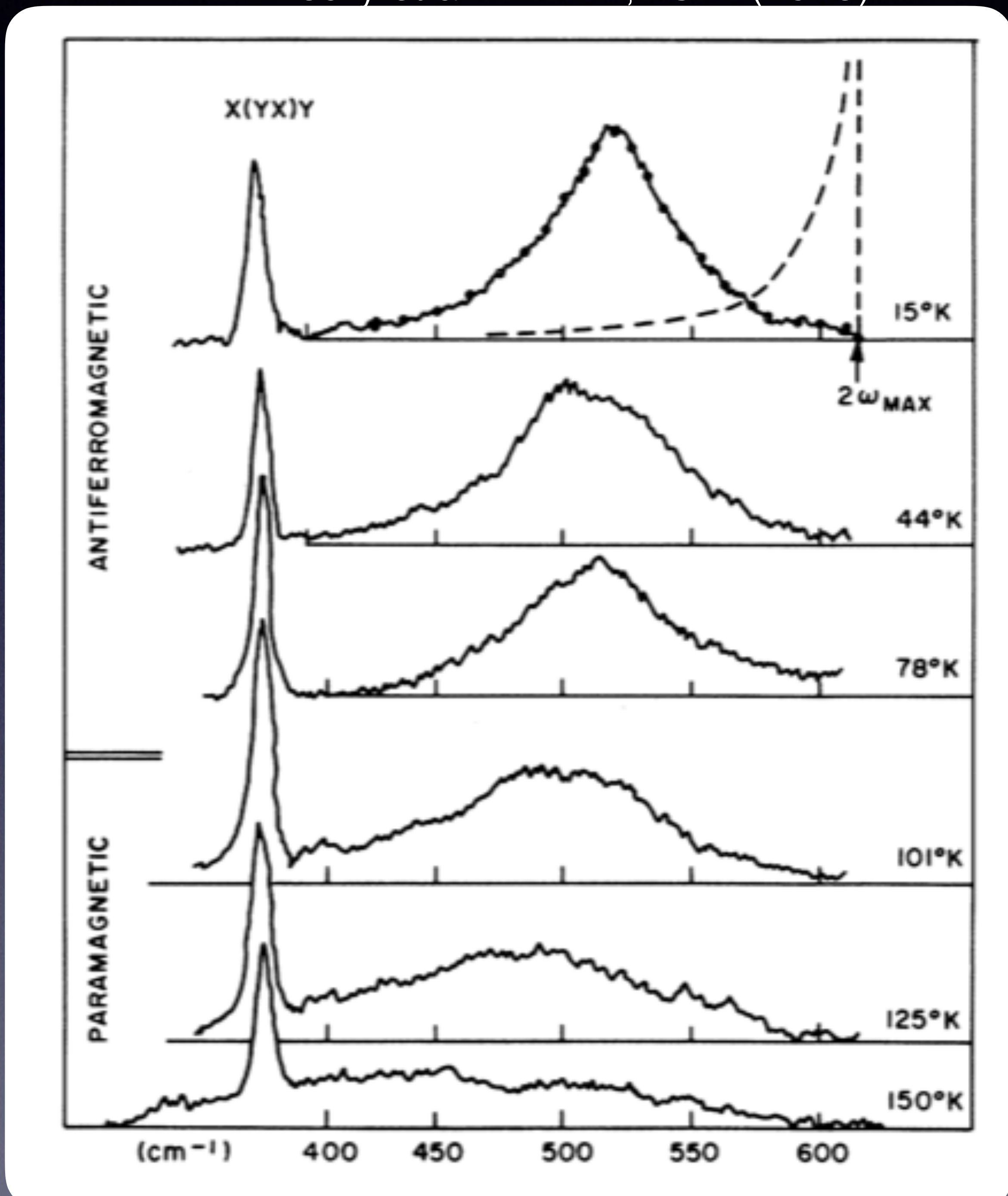


- K_2NiF_4, T_N = 96 K
- 2M-mode up to T ~ 1.5 x T_N

Long-range order

Length-scale dynamics

P. Fleury *et al.* PRL **24**, 1347 (1970)



- K_2NiF_4, T_N = 96 K
- 2M-mode up to T ~ 1.5 x T_N

Long-range order

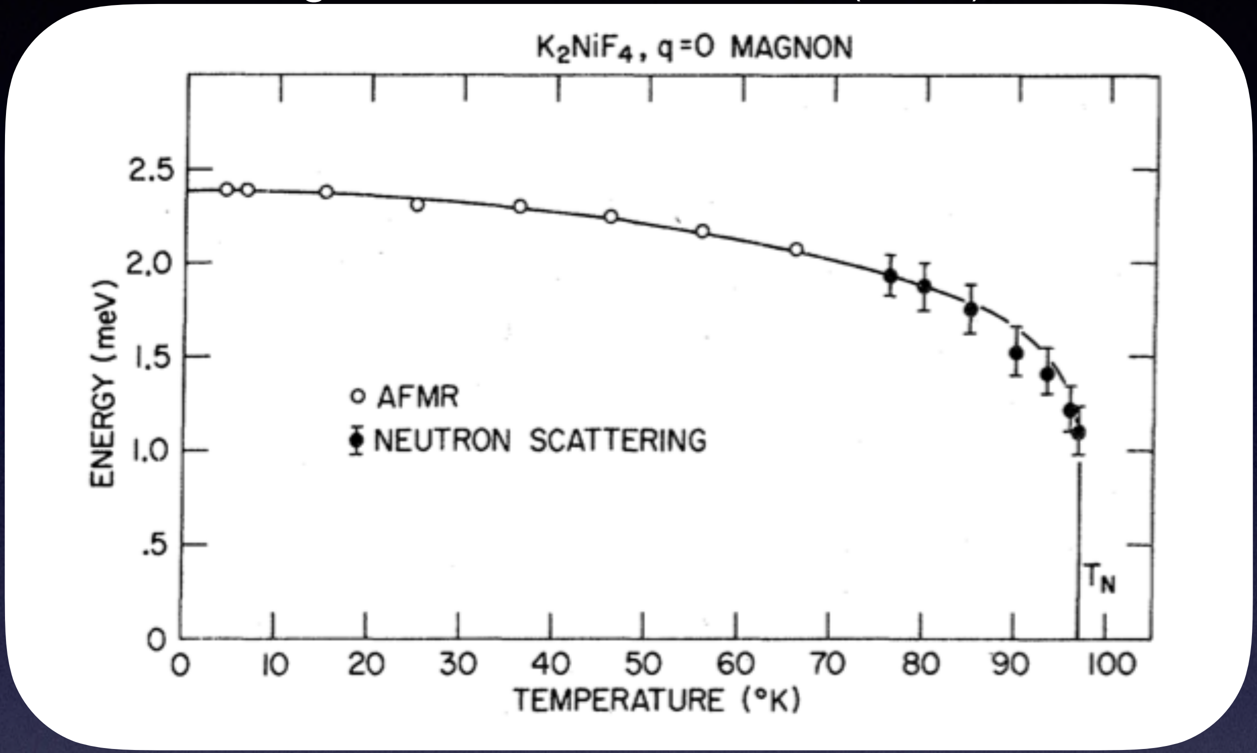
T

T_N

Short-range spin-spin correlations

Length-scale dynamics

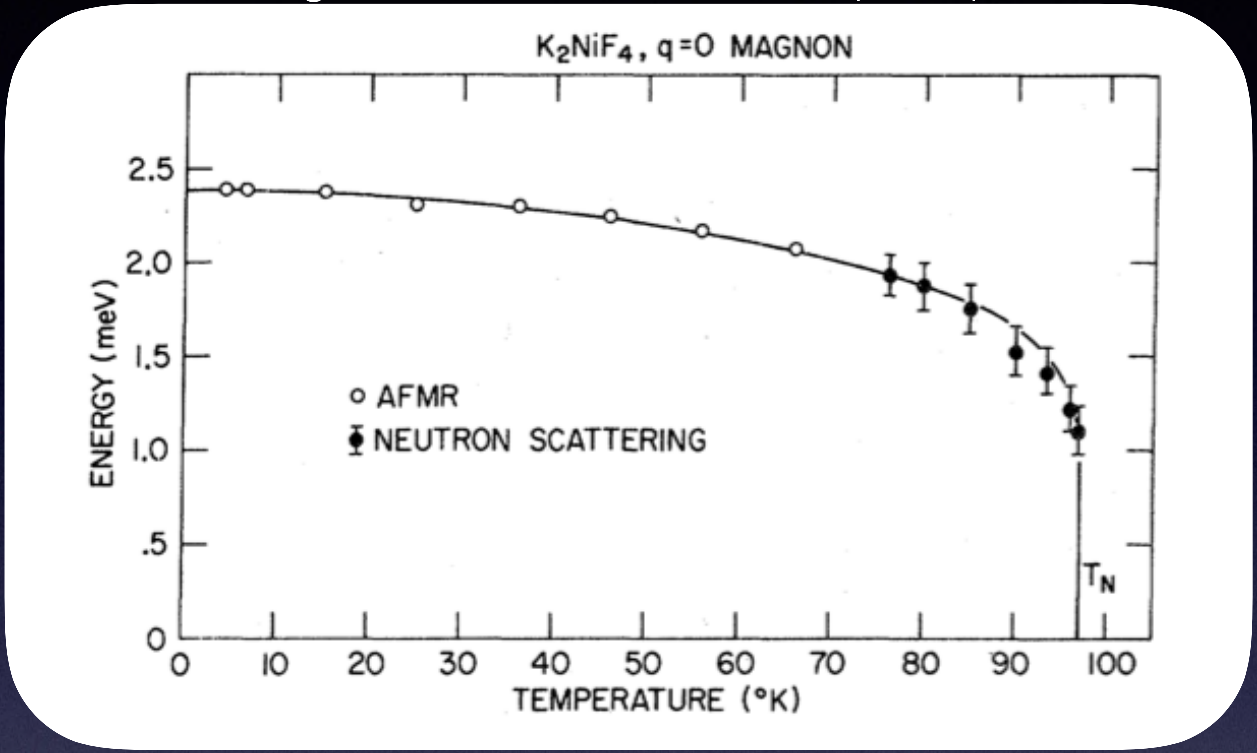
R. Birgenau *et al.* PRB **3**, 1736 (1971)



k~0 magnons soften at T_N

Length-scale dynamics

R. Birgenau *et al.* PRB **3**, 1736 (1971)

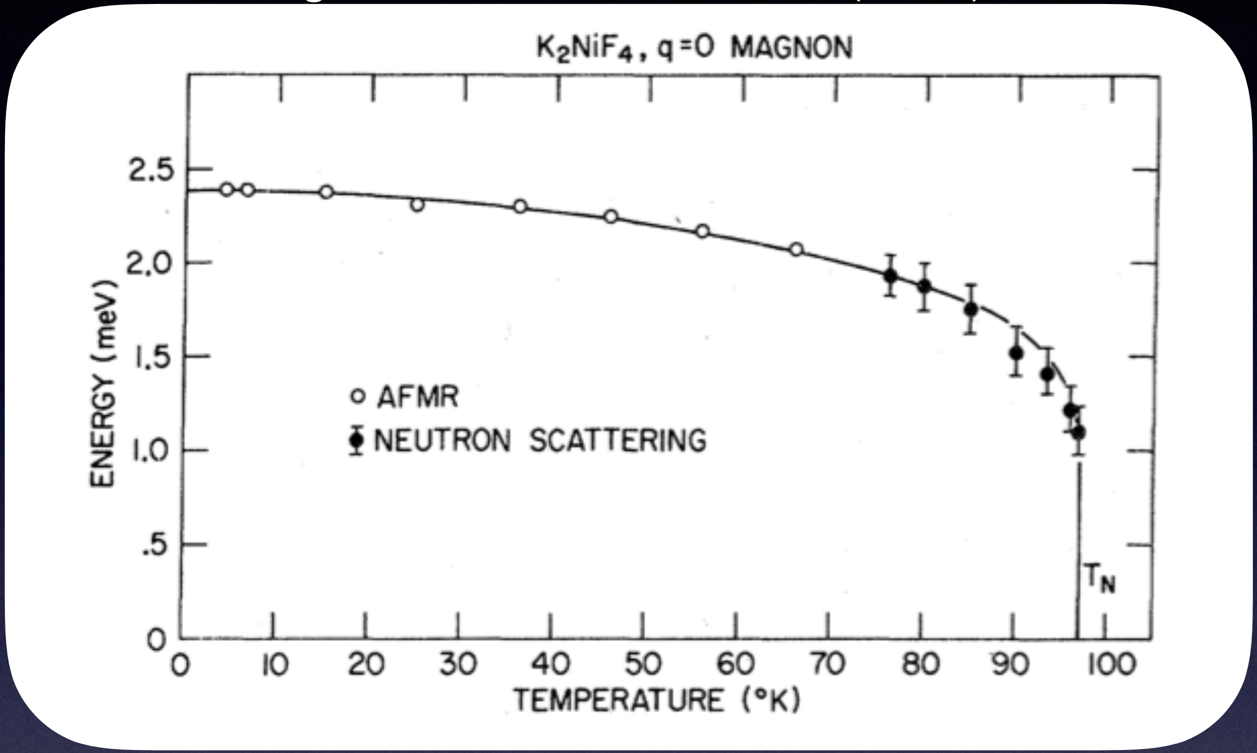


k~0 magnons soften at T_N

Time-domain experiments no signal above T_N

Length-scale dynamics

R. Birgenau *et al.* PRB **3**, 1736 (1971)



k~0 magnons soften at T_N

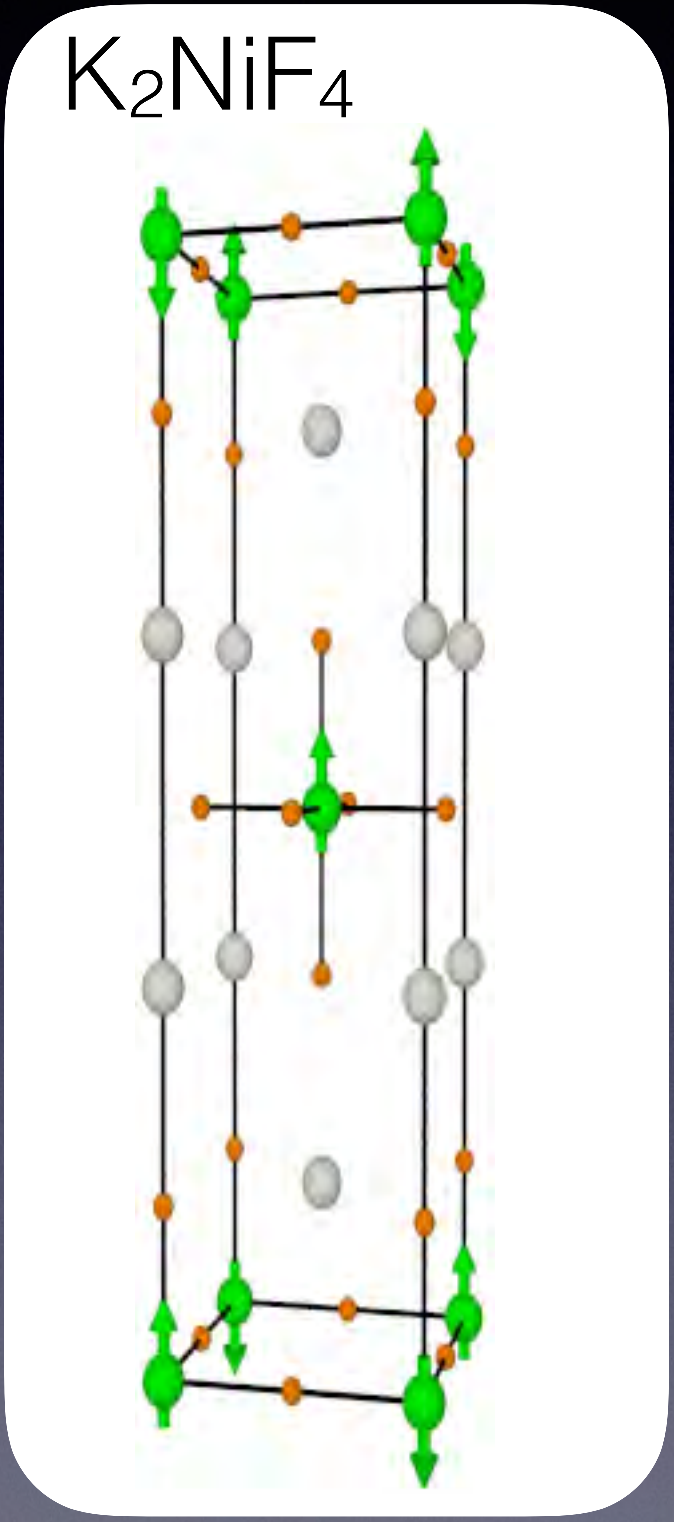
Time-domain experiments no signal above T_N

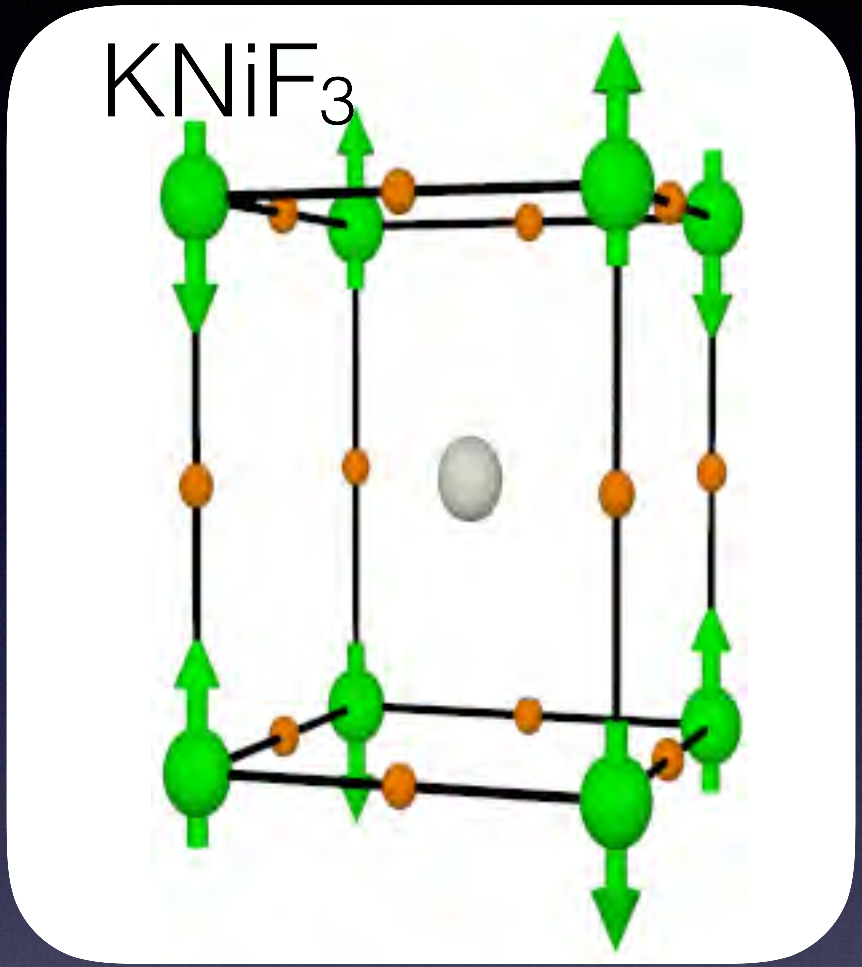
J. Zhao *et al.* PRB 7**3**, 184434 (2006)

Long-wavelength magnons contributions ?

Our approach

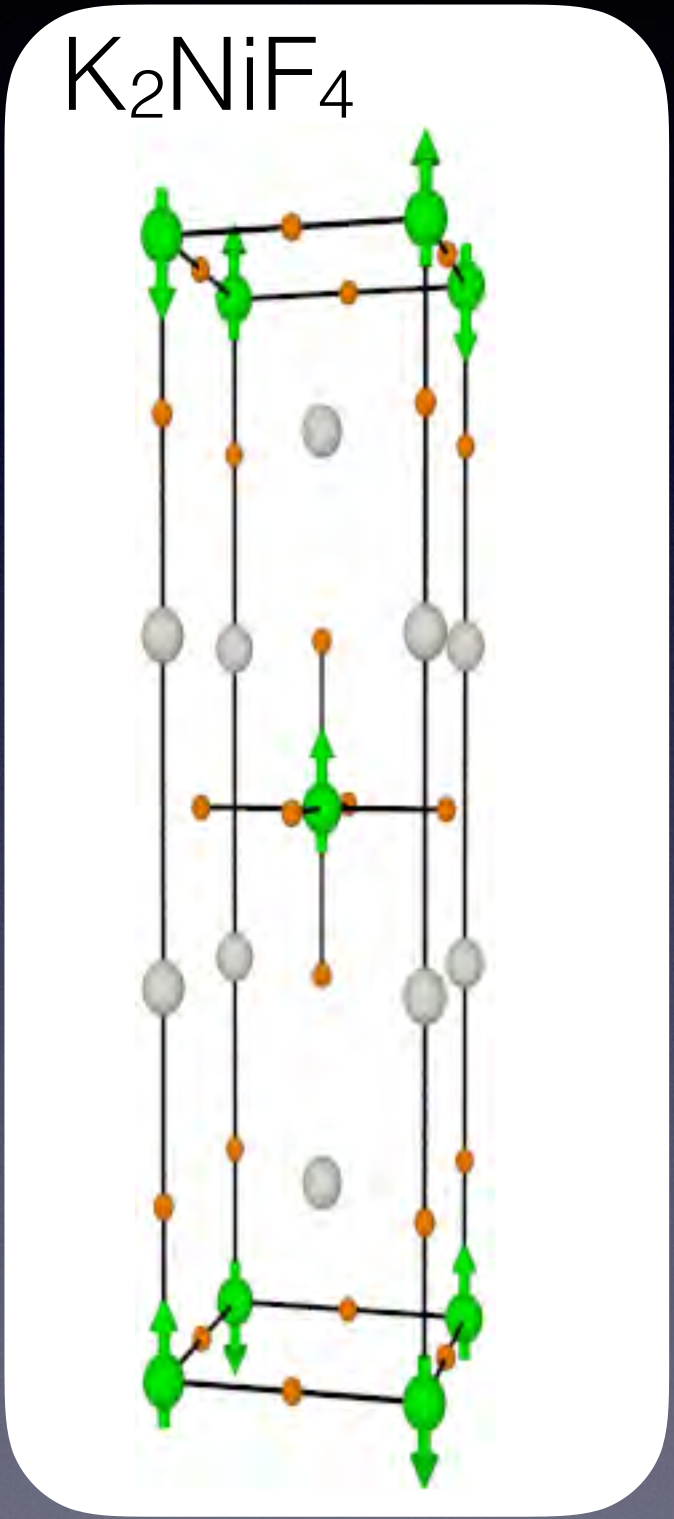
Our approach

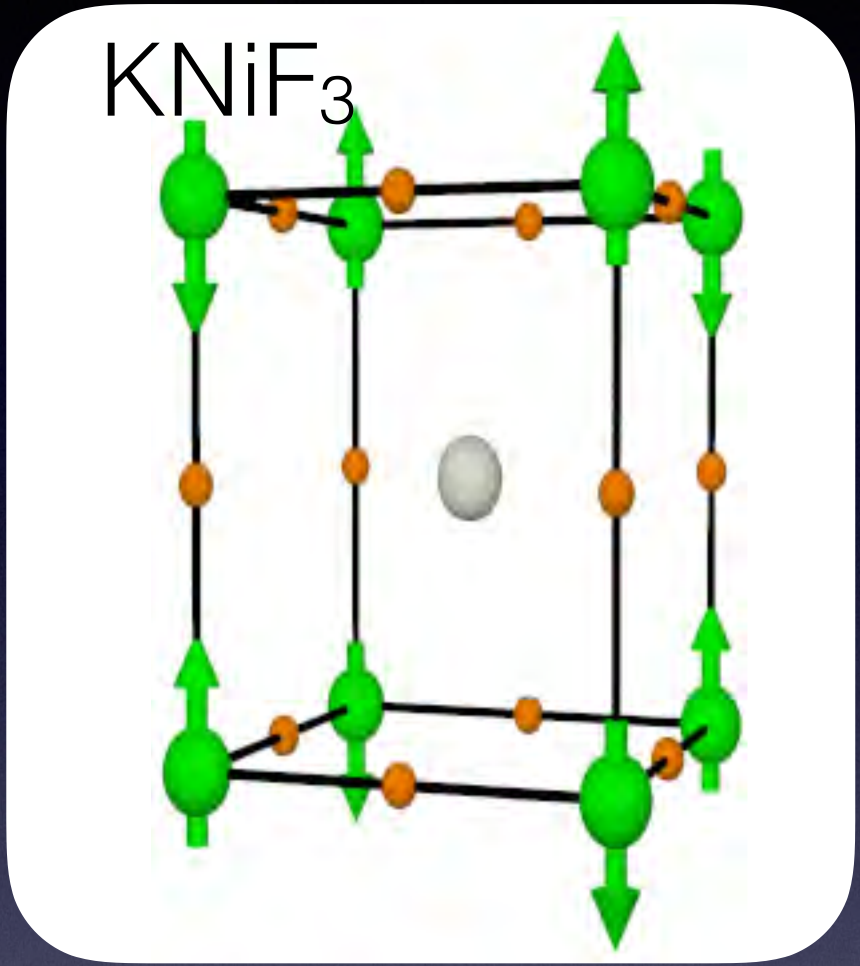




- same exchange (short-range)
- different anisotropy (long-range)

Our approach

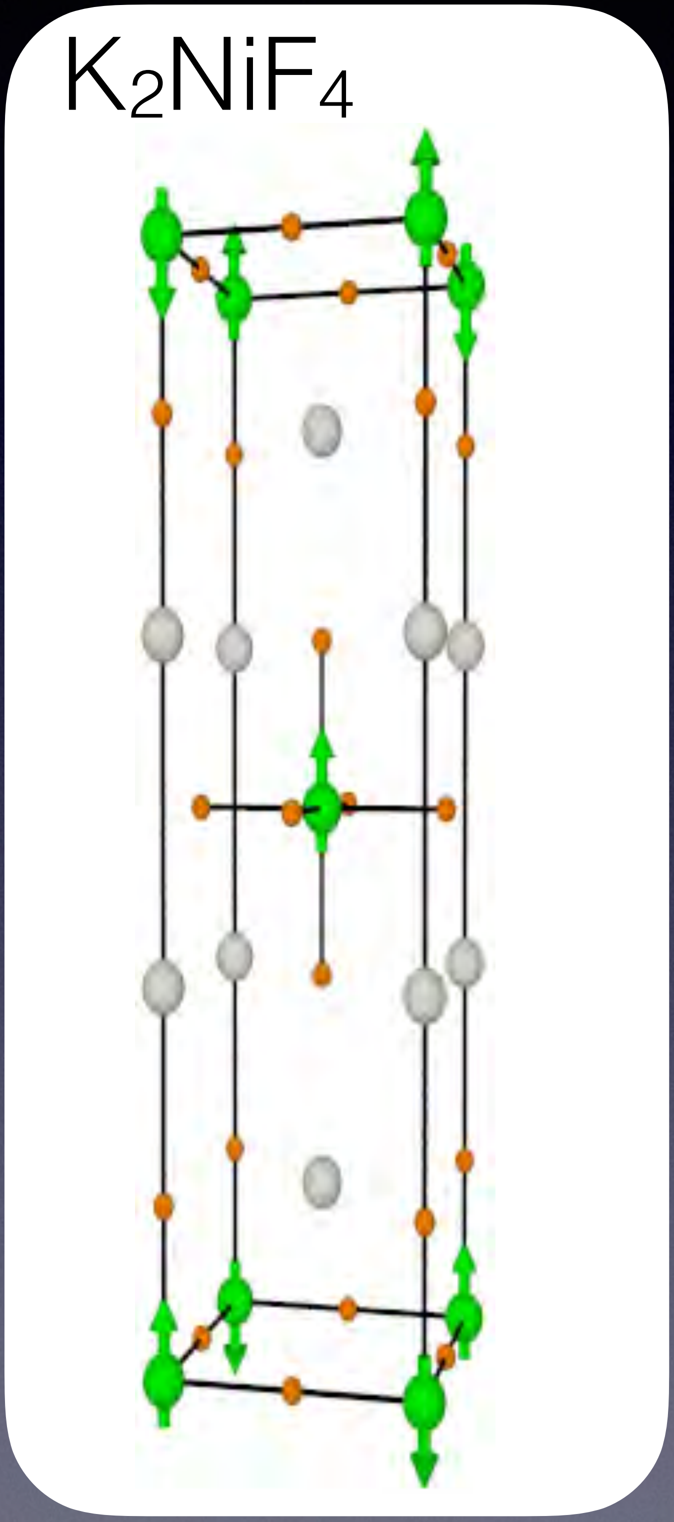


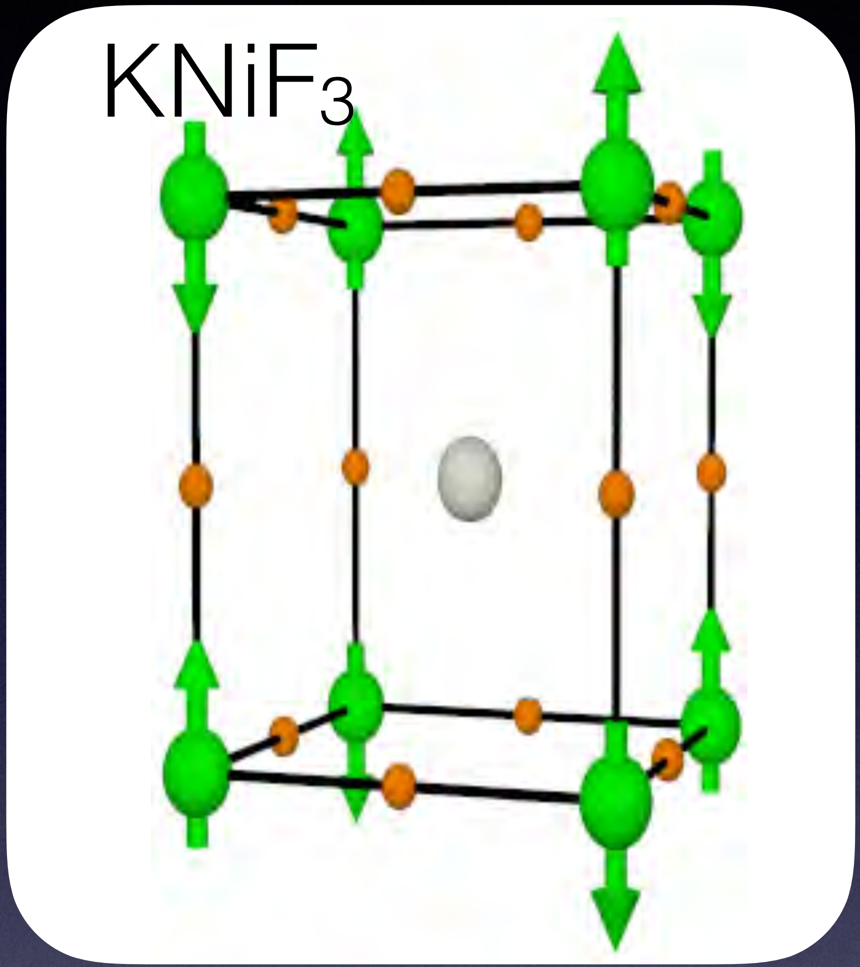


- same exchange (short-range)
- different anisotropy (long-range)

Temperature dependence
femto-nanomagnonics

Our approach



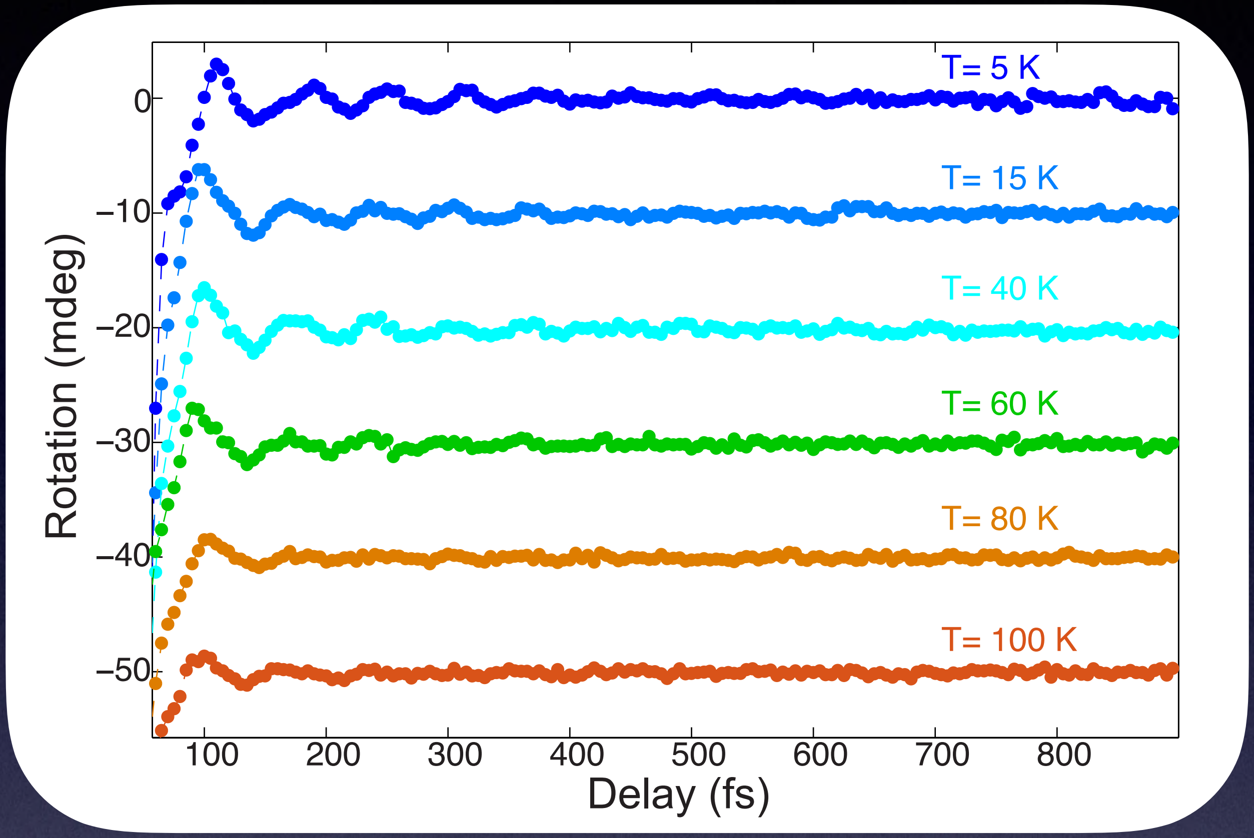


- same exchange (short-range)
- different anisotropy (long-range)

Temperature dependence femto-nanomagnonics

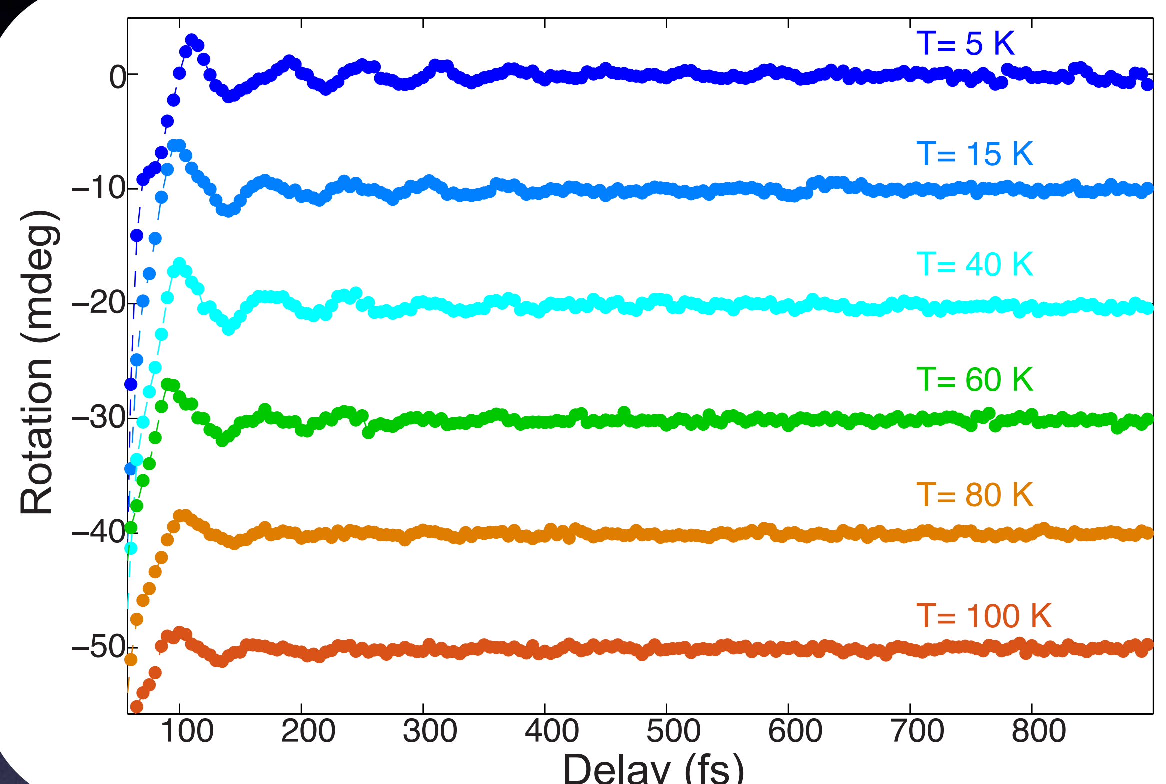
Softening and/or divergence evidence of long-range

Temperature dependence

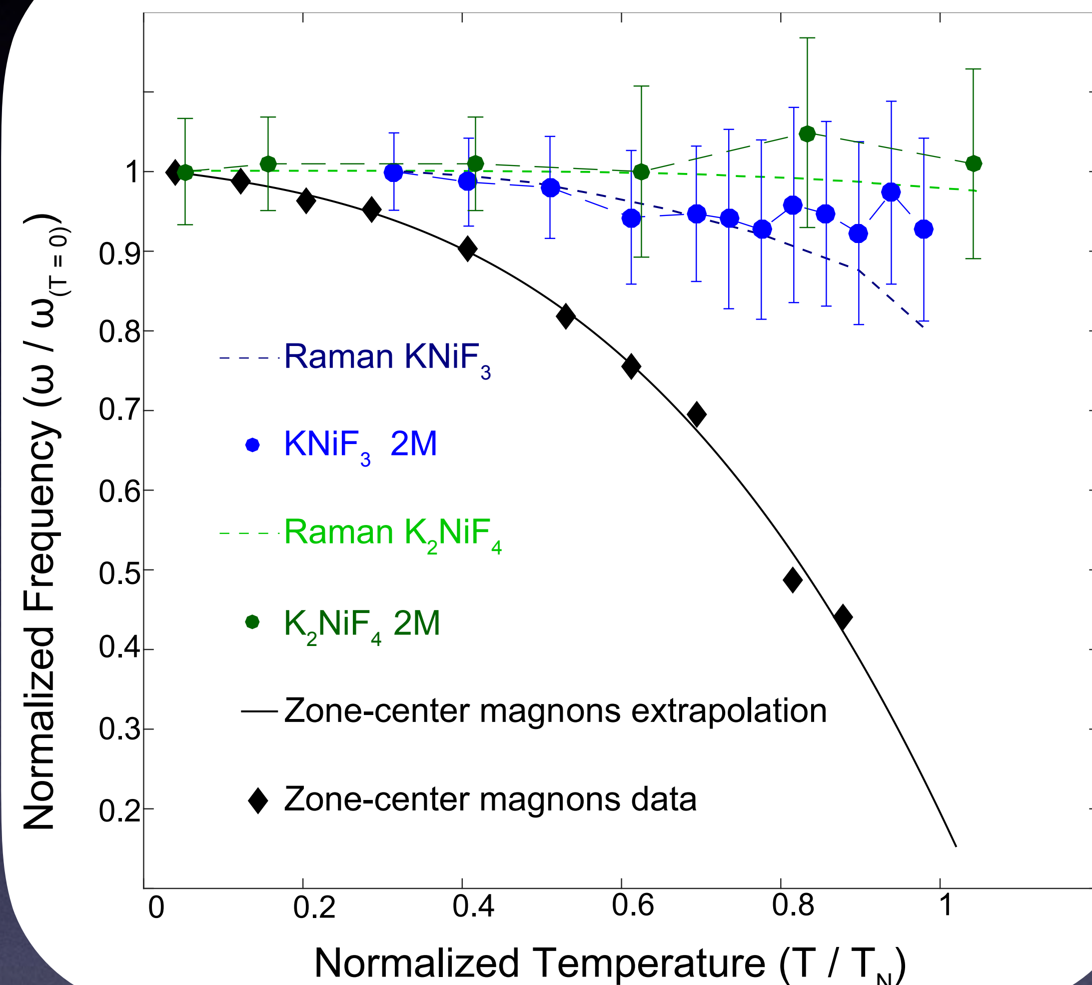


- Pump: 1.9 eV, Probe = 1.3 eV
- Fluence = 4.5 mJ/cm^2

Temperature dependence

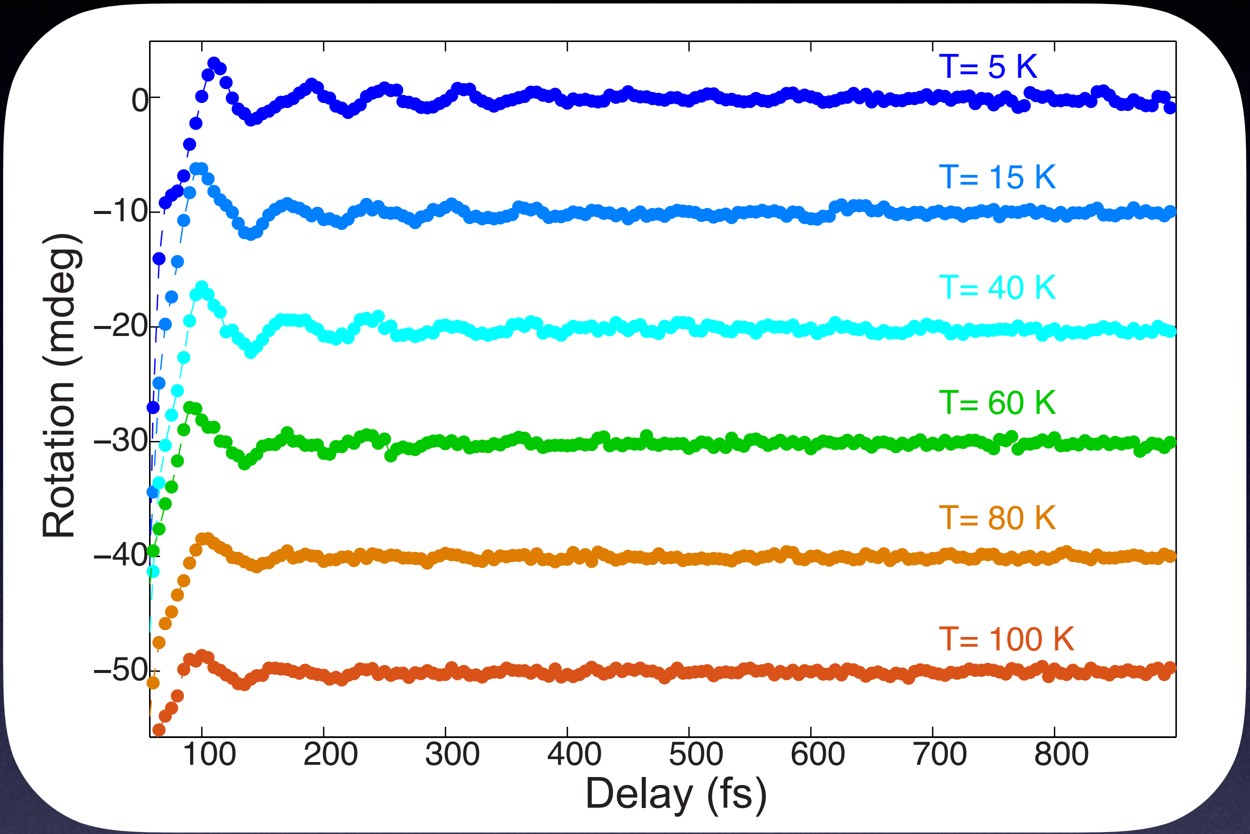


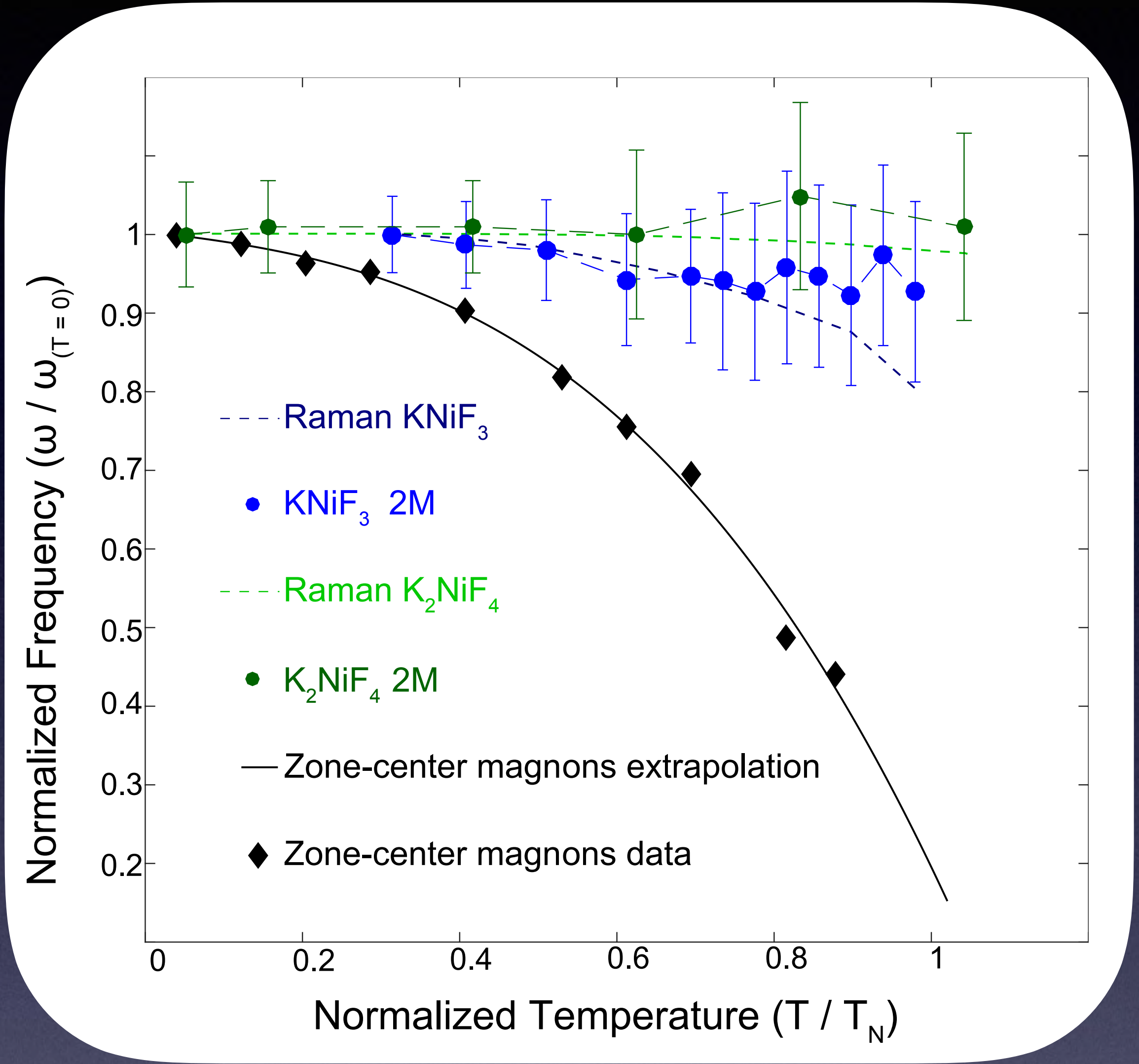
- Pump: 1.9 eV, Probe = 1.3 eV
- Fluence = 4.5 mJ/cm²



D. Bossini *et al.* in preparation

Temperature dependence





D. Bossini *et al.* in preparation

- Pump: 1.9 eV, Probe = 1.3 eV
- Fluence = 4.5 mJ/cm^2

Contribution only from femto-nanomagnons

Role of temperature

Temperature defines the amplitude and lifetime
(magnon-magnon interaction)

S. Chinn et al. PRB **3**, 1709 (1971)

U. Balucani et al. PRB **8**, 4247 (1973)

Role of temperature

Temperature defines the amplitude and lifetime (magnon-magnon interaction)

S. Chinn et al. PRB **3**, 1709 (1971)

U. Balucani et al. PRB **8**, 4247 (1973)

Softening temperature (NO T_N): massive thermal population of femto-nanomagnons

Role of temperature

Temperature defines the amplitude and lifetime (magnon-magnon interaction)

S. Chinn et al. PRB **3**, 1709 (1971)

U. Balucani et al. PRB **8**, 4247 (1973)

Softening temperature (NO T_N): massive thermal population of femto-nanomagnons

$KNiF_3$ $T_s \approx 1.2 \cdot 10^{-20}\,\mathrm{J}$ $T_N \approx 0.3 \cdot 10^{-20}\,\mathrm{J}$

Role of temperature

Temperature defines the amplitude and lifetime (magnon-magnon interaction)

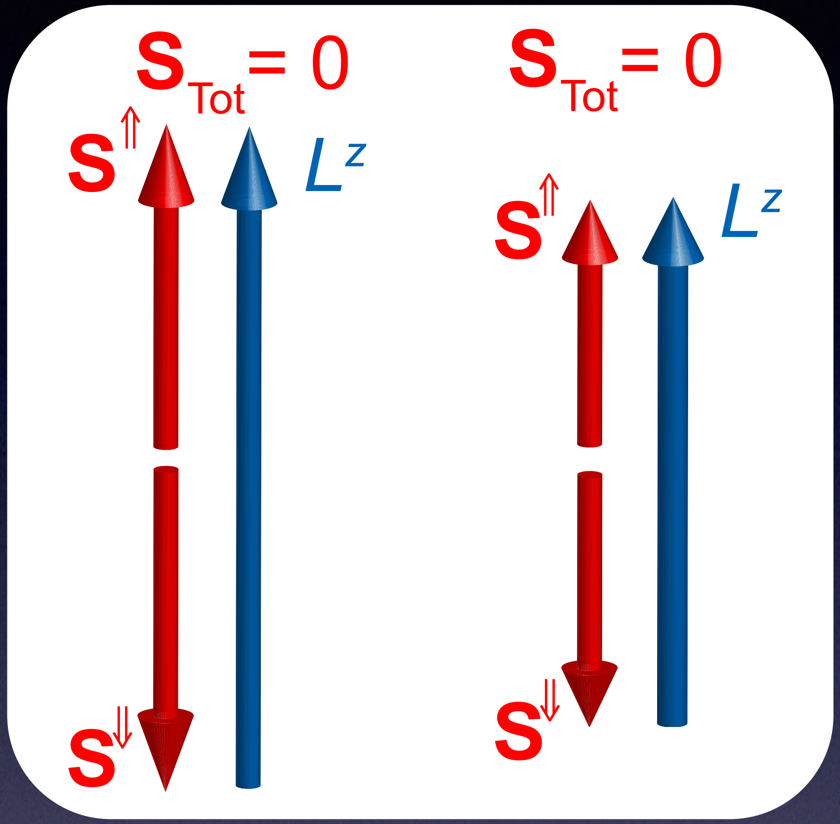
S. Chinn et al. PRB **3**, 1709 (1971)

U. Balucani et al. PRB **8**, 4247 (1973)

Softening temperature (NO T_N): massive thermal population of femto-nanomagnons

$KNiF_3$	$T_s \approx 1.2 \cdot 10^{-20}\mathrm{J}$	$T_N \approx 0.3 \cdot 10^{-20}\mathrm{J}$
K_2NiF_4	$T_s \approx 1 \cdot 10^{-20}\mathrm{J}$	$T_N \approx 0.1 \cdot 10^{-20}\mathrm{J}$

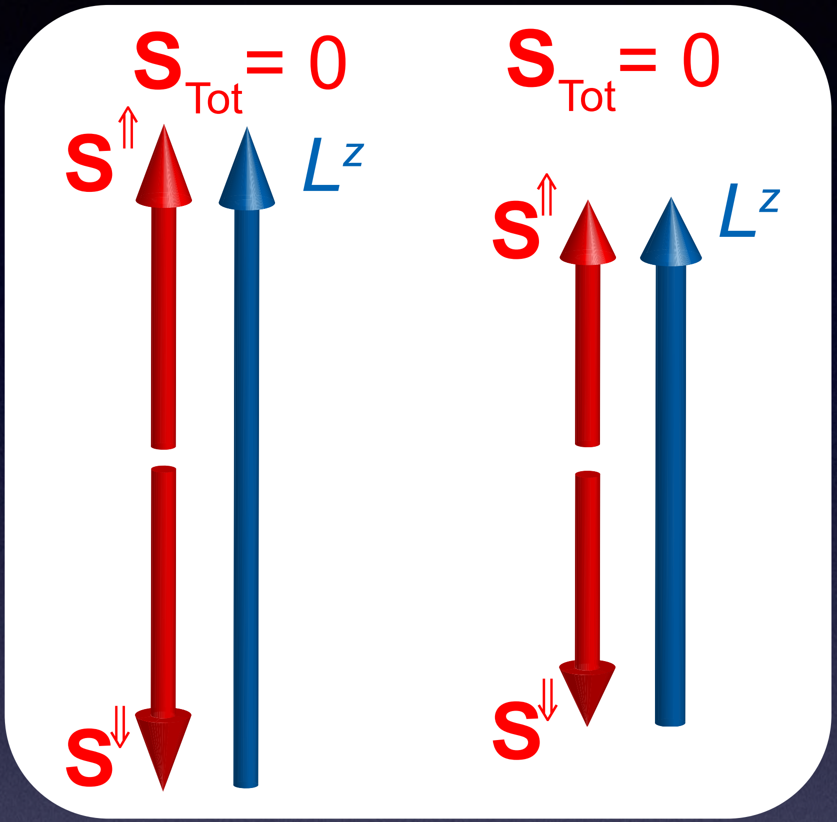
Dynamics order parameter



$\Delta S = 0$

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

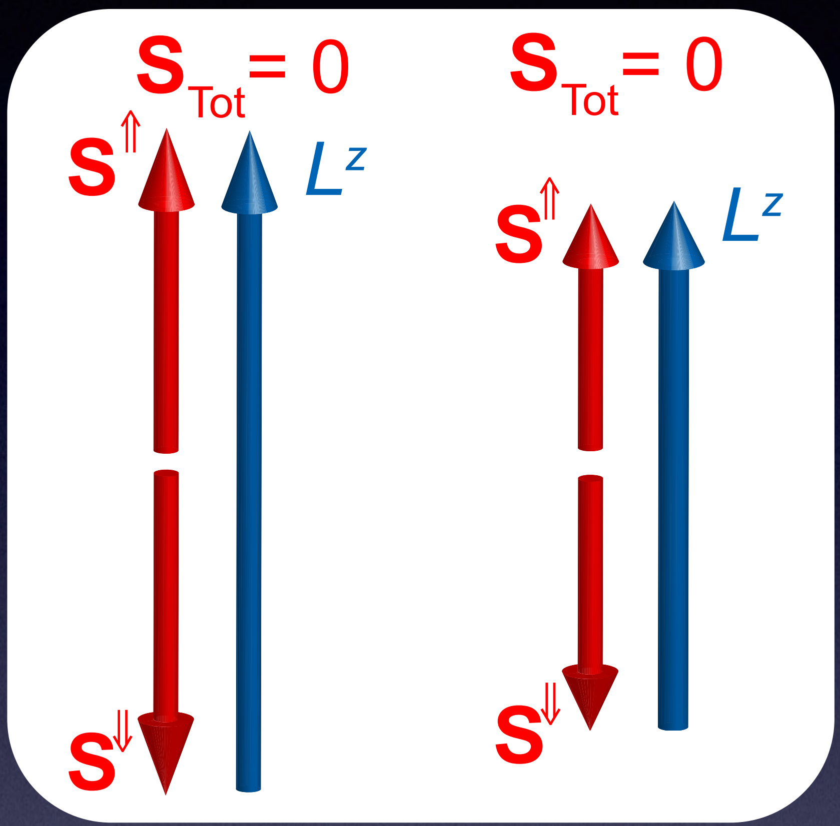
Dynamics order parameter



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

$\Delta S = 0$ No magnetization dynamics (no angular momentum)

Dynamics order parameter

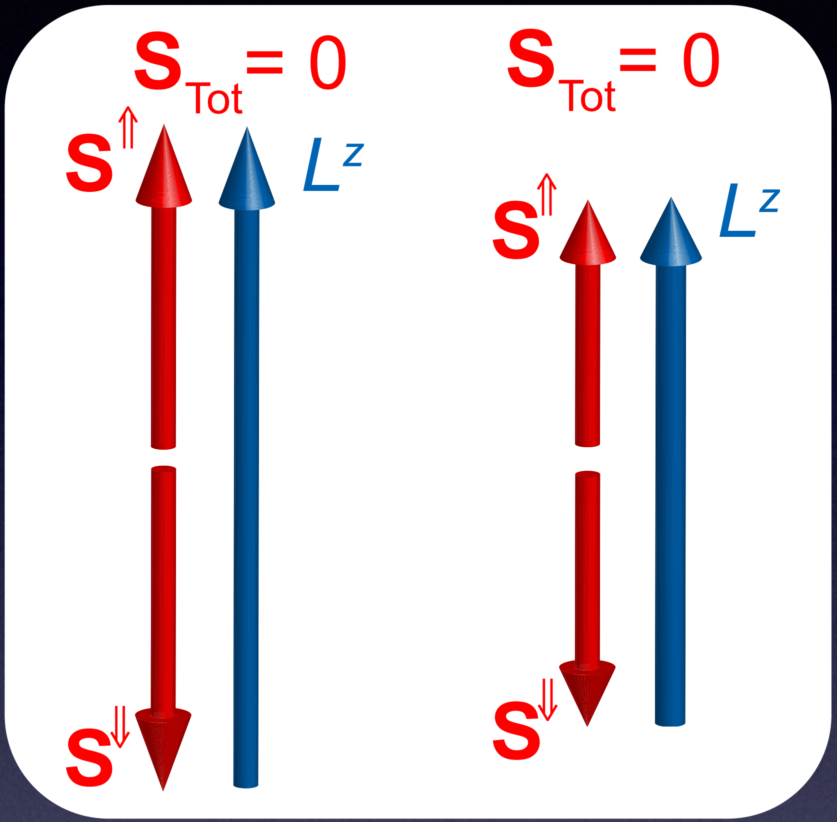


$$L \equiv S^{\Uparrow} - S^{\Downarrow}$$

$\Delta S = 0$ No magnetization dynamics (no angular momentum)

Precession forbidden, dynamics purely longitudinal!

Dynamics order parameter



ΔS = 0 No magnetization dynamics (no angular momentum)

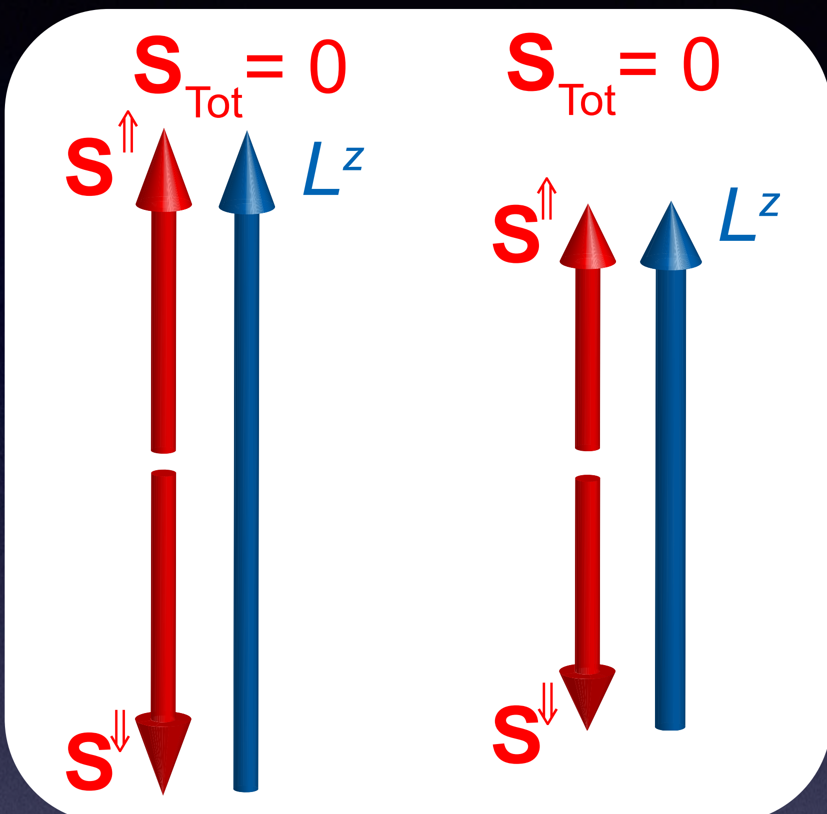
Precession forbidden, dynamics purely longitudinal!

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

J. Zhao et al. PRL **93**, 107203 (2004)

$$\hat{H}_1(t) = \delta(t)\frac{4\pi I_1}{n_R c}\sum_{\langle i,j\rangle}\Xi_{ij}\left(\frac{\hat{S}_i^{+\Uparrow}\hat{S}_j^{-\Downarrow} + \hat{S}_i^{-\Uparrow}\hat{S}_j^{+\Downarrow}}{2} + A\hat{S}_i^{z\Uparrow}\hat{S}_j^{z\Downarrow}\right)$$

Dynamics order parameter



ΔS = 0 No magnetization dynamics (no angular momentum)

Precession forbidden, dynamics purely longitudinal!

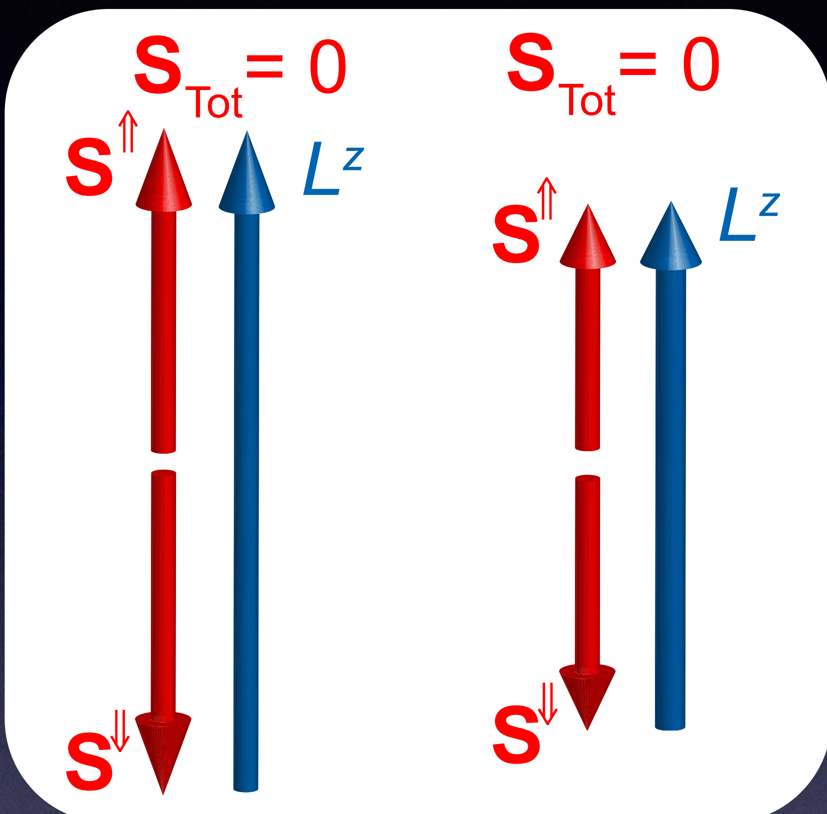
$$\boldsymbol{L} \equiv \boldsymbol{S}^{\Uparrow} - \boldsymbol{S}^{\Downarrow}$$

J. Zhao et al. PRL **93**, 107203 (2004)

$$\hat{H}_1(t) = \delta(t)\frac{4\pi I_1}{n_R c}\sum_{\langle i,j\rangle}\Xi_{ij}\left(\frac{\hat{S}_i^{+\Uparrow}\hat{S}_j^{-\Downarrow} + \hat{S}_i^{-\Uparrow}\hat{S}_j^{+\Downarrow}}{2} + A\hat{S}_i^{z\Uparrow}\hat{S}_j^{z\Downarrow}\right)$$

Symmetric in x-y plane

Dynamics order parameter



ΔS = 0 No magnetization dynamics
(no angular momentum)

Precession forbidden, dynamics purely longitudinal!

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

J. Zhao et al. PRL **93**, 107203 (2004)

$$\hat{H}_1(t) = \delta(t)\frac{4\pi I_1}{n_R c}\sum_{\langle i,j\rangle}\Xi_{ij}\left(\frac{\hat{S}_i^{+\Uparrow}\hat{S}_j^{-\Downarrow}+\hat{S}_i^{-\Uparrow}\hat{S}_j^{+\Downarrow}}{2}+A\hat{S}_i^{z\Uparrow}\hat{S}_j^{z\Downarrow}\right)$$

Symmetric in x-y plane

Thermodynamics?

Thermodynamics

$$\mathrm{M} = \mathrm{M}(\mathrm{T})$$

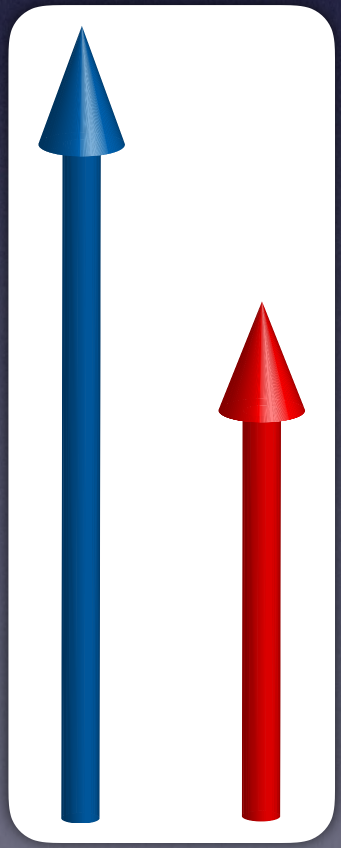
$$\mathrm{L} = \mathrm{L}(\mathrm{T})$$

Thermodynamics?

Thermodynamics

$$\mathrm{M} = \mathrm{M}(\mathrm{T})$$

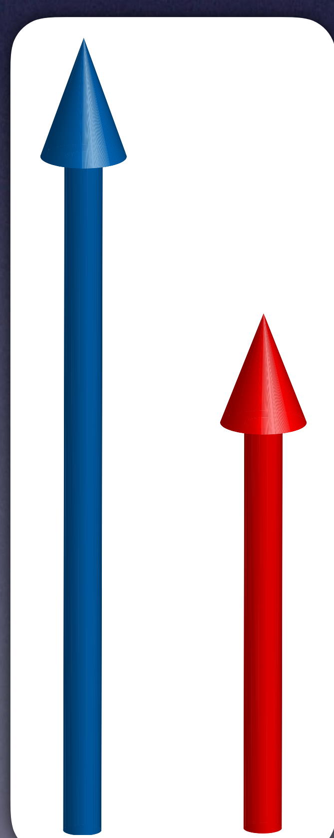
$$\mathrm{L} = \mathrm{L}(\mathrm{T})$$

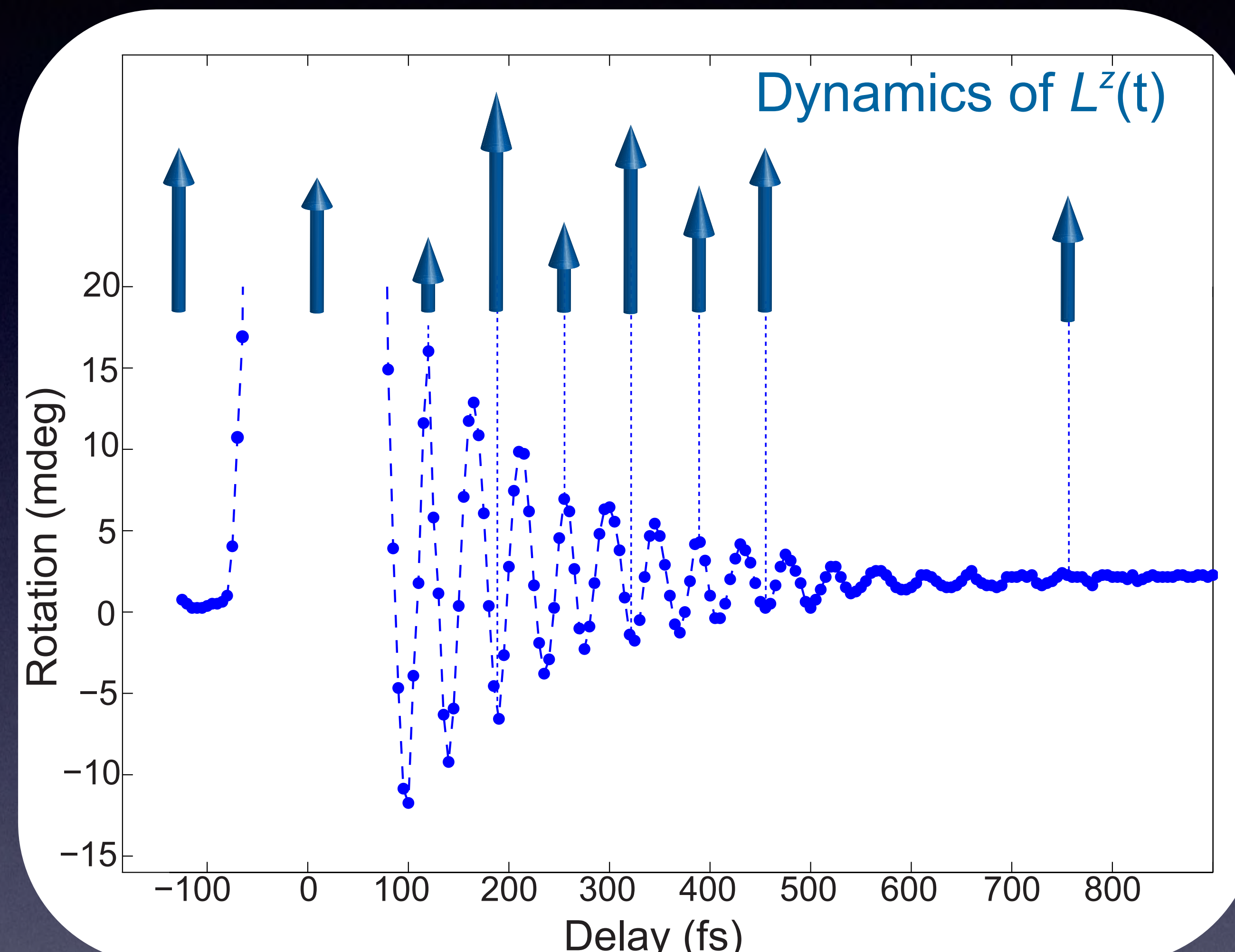


Thermodynamics?

Thermodynamics

$$M = M(T)$$
$$L = L(T)$$

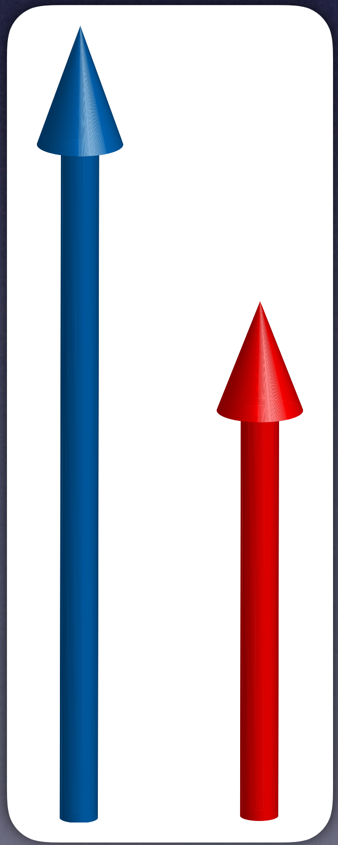


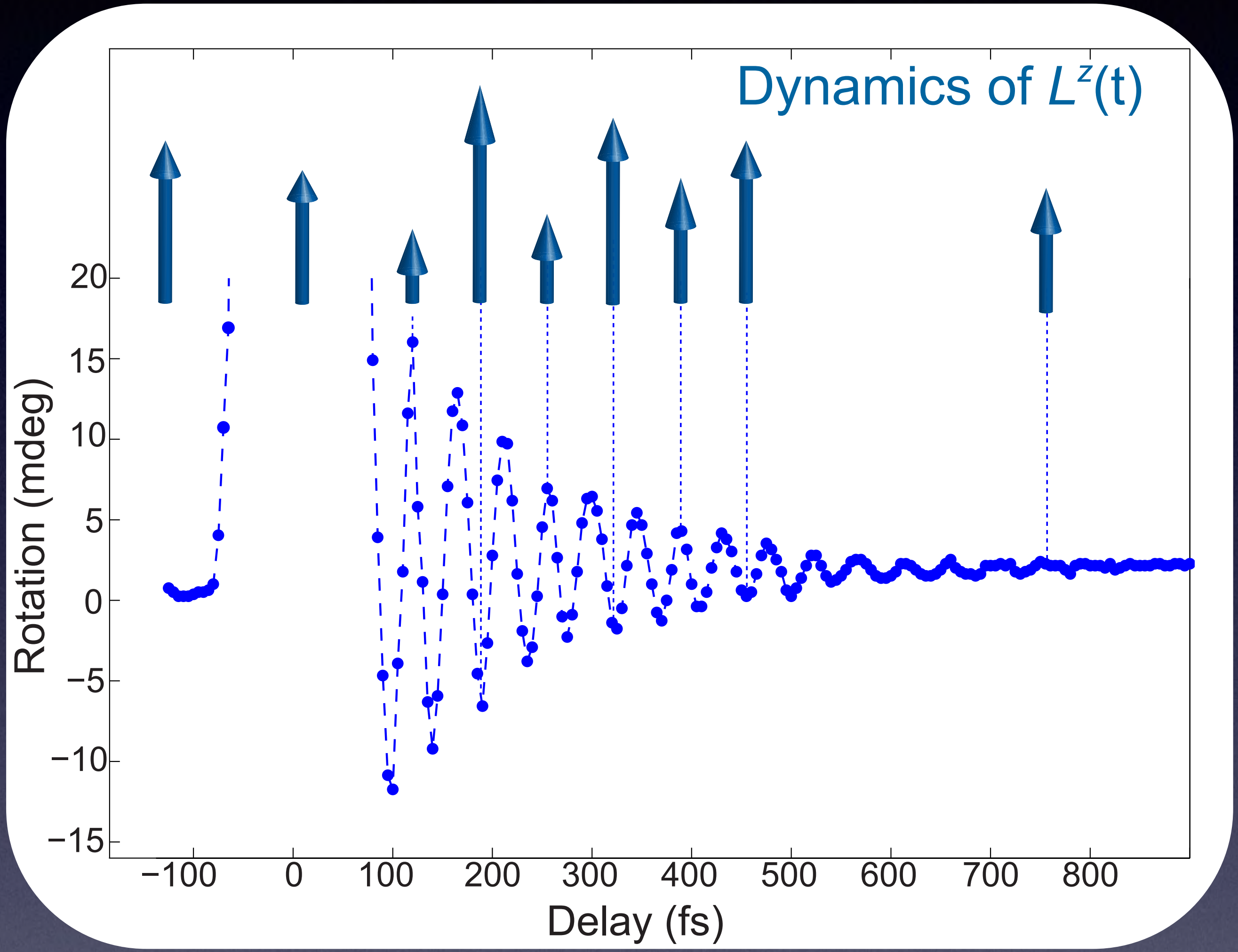


Thermodynamics?

Thermodynamics

$$M = M(T)$$
$$L = L(T)$$





Oscillations of T coherently controlled

Thermodynamics?

Further reasons: effective field, equation of motion

Thermodynamics?

Further reasons: effective field, equation of motion

Classically: mean field approach to spin dynamics (precession)

Thermodynamics?

Further reasons: effective field, equation of motion

Classically: mean field approach to spin dynamics (precession)

$$\langle \boldsymbol{S}_i \boldsymbol{S}_j \rangle = \langle \boldsymbol{S}_i \rangle \langle \boldsymbol{S}_j \rangle$$

Thermodynamics?

Further reasons: effective field, equation of motion

Classically: mean field approach to spin dynamics (precession)

$$\langle \boldsymbol{S}_i \boldsymbol{S}_j \rangle = \langle \boldsymbol{S}_i \rangle \langle \boldsymbol{S}_j \rangle$$

Femto-nanomagnonics: dynamics of nearest neighbors correlations

Thermodynamics?

Further reasons: effective field, equation of motion

Classically: mean field approach to spin dynamics (precession)

$$\langle \boldsymbol{S}_i \boldsymbol{S}_j \rangle = \langle \boldsymbol{S}_i \rangle \langle \boldsymbol{S}_j \rangle$$

Femto-nanomagnonics: dynamics of nearest neighbors correlations

Quantum modelling

Goal: equation of motion

Quantum modelling

Goal: equation of motion

$$H_0 = J \sum_{i,\boldsymbol{\delta}} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+\boldsymbol{\delta}} \qquad \delta H = \frac{1}{2} f(t) \sum_{i,\boldsymbol{\delta}} \Delta J(\boldsymbol{\delta}) \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+\boldsymbol{\delta}},$$

Quantum modelling

Goal: equation of motion

$$H_0 = J \sum_{i,\boldsymbol{\delta}} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+\boldsymbol{\delta}} \qquad \delta H = \frac{1}{2} f(t) \sum_{i,\boldsymbol{\delta}} \Delta J(\boldsymbol{\delta}) \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+\boldsymbol{\delta}},$$

$$|\mu_{\mathbf{k}}\rangle = \sqrt{1 - |\mu_{\mathbf{k}}|^2} \sum_{n=0}^{\infty} \mu_{\mathbf{k}}^n |n_{\mathbf{k}}\rangle |n_{\mathbf{k}}\rangle$$

Quantum modelling

Goal: equation of motion

$$H_0 = J\sum_{i,\boldsymbol{\delta}} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+\boldsymbol{\delta}} \qquad \delta H = \frac{1}{2} f(t) \sum_{i,\boldsymbol{\delta}} \Delta J(\boldsymbol{\delta}) \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+\boldsymbol{\delta}},$$

$$|\mu_{\mathbf{k}}\rangle = \sqrt{1-|\mu_{\mathbf{k}}|^2} \sum_{n=0}^{\infty} \mu_{\mathbf{k}}^n |n_{\mathbf{k}}\rangle |n_{\mathbf{k}}\rangle \qquad \partial_t \mu_{\mathbf{k}} = \{\mu_{\mathbf{k}}, H(\mu_{\mathbf{k}}, \mu_{\mathbf{k}}^*)\}$$

Quantum modelling

Goal: equation of motion

$$H_0 = J \sum_{i,\boldsymbol{\delta}} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+\boldsymbol{\delta}} \qquad \delta H = \frac{1}{2} f(t) \sum_{i,\boldsymbol{\delta}} \Delta J(\boldsymbol{\delta}) \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+\boldsymbol{\delta}},$$

$$|\mu_{\mathbf{k}}\rangle = \sqrt{1 - |\mu_{\mathbf{k}}|^2} \sum_{n=0}^{\infty} \mu_{\mathbf{k}}^n |n_{\mathbf{k}}\rangle |n_{\mathbf{k}}\rangle \qquad \partial_t \mu_{\mathbf{k}} = \{\mu_{\mathbf{k}}, H(\mu_{\mathbf{k}}, \mu_{\mathbf{k}}^*)\}$$

$$L_z = L_z(0) - zS \sum_{\mathbf{k}} \frac{\gamma_{\mathbf{k}}}{\sqrt{1 - \gamma_{\mathbf{k}}^2}} \frac{2\mathrm{Re}\mu_{\mathbf{k}}}{1 - |\mu_{\mathbf{k}}|^2}$$

D. Bossini *et al.* in preparation

What's next

Femto-nanomagnonics

What's next

Femto-nanomagnonics

✓ Coherent control (ISRS excitation)

What's next

Femto-nanomagnonics

- ✓ Coherent control (ISRS excitation)
- ☐ Manipulate magnetic phases via femto-nanomagnons

What's next

Femto-nanomagnonics

✓ Coherent control (ISRS excitation)

☐ Manipulate magnetic phases via femto-nanomagnons

Resonant pumping ?

What's next

Femto-nanomagnonics

- ✓ Coherent control (ISRS excitation)
- ☐ Manipulate magnetic phases via femto-nanomagnons

Resonant pumping ?

- Direct IR-pumping

What’s next

Femto-nanomagnonics

- ✓ Coherent control (ISRS excitation)
- ☐ Manipulate magnetic phases via femto-nanomagnons

Resonant pumping ?

- Direct IR-pumping
- Phonon-assisted IR-pumping

What's next

Femto-nanomagnonics

- ✓ Coherent control (ISRS excitation)
- ☐ Manipulate magnetic phases via femto-nanomagnons

Resonant pumping ?

- Direct IR-pumping
- Phonon-assisted IR-pumping
- Exciton-magnon process (visible-nearIR)

What's next

Femto-nanomagnonics

- ✓ Coherent control (ISRS excitation)
- ☐ Manipulate magnetic phases via femto-nanomagnons

Resonant pumping ?

- Direct IR-pumping
- Phonon-assisted IR-pumping
- Exciton-magnon process (visible-nearIR)

D. Bossini *et al.* in preparation

Conclusions

1. Excitation, control and detection of femto-nanomagnons
2. Disclosure of quantum spin dynamics

Conclusions

1. Excitation, control and detection of femto-nanomagnons
2. Disclosure of quantum spin dynamics

Femto-nanomagnonics!

D. Bossini et al. Nat. Comm. **7,** *10645 (2016)*

D. Bossini et al. Physica Scritpa **92**, 024002 (2017)

Conclusions

1. Excitation, control and detection of femto-nanomagnons
2. Disclosure of quantum spin dynamics

Femto-nanomagnonics!

D. Bossini et al. Nat. Comm. **7,** *10645 (2016)*

D. Bossini et al. Physica Scritpa **92**, 024002 (2017)

- [ ] Experimental prove of quantum nature
- [ ] Spatial propagation

Acknowledgements

S. Dal Conte, G. Cerullo
IFN-CNR Dipartimento di Fisica, Politecnico di Milano, Milano, Italy

R. V. Pisarev
Ioffe Physical-Technical Institute, Russian Academy of Sciences, St. Petersburg, Russia

A. Secchi, Y. Hashimoto, J. Mentink, Th. Rasing, A.V. Kimel
Radboud University Nijmegen, Institute for Molecules and Materials, Nijmegen, The Netherlands

H. Gomonay, J. Sinova
Institut für Physik, Johannes Gutenberg Universität Mainz, 55128 Mainz, Germany

D. Mihailovic
Jozef Stefan Institute & CENN-Nanocenter, Jamova 39, Ljubljana SI-1000, Slovenia