Femtosecond dynamics of antiferromagnets and entangled magnon states

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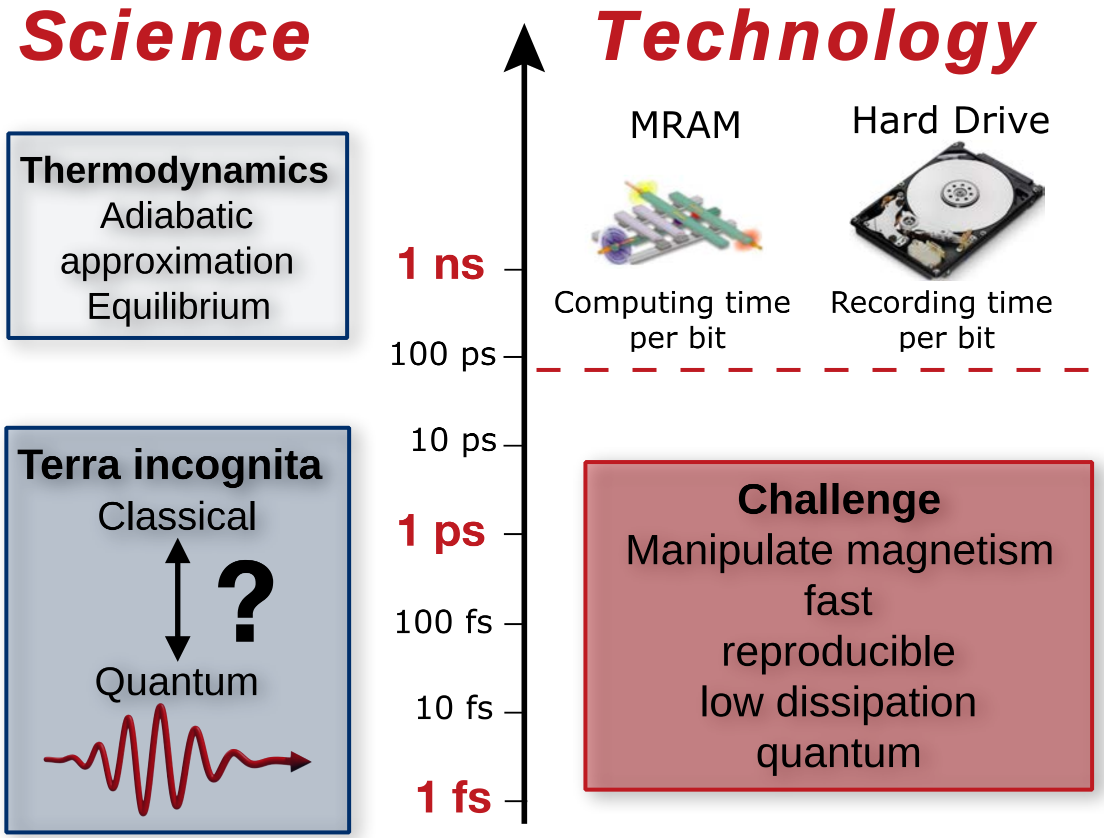
Ultrafast Spintronics Workshop

Take-home message

- Short-range correlations in AF + light ⇒ **macroscopic quantum** 2-magnon states
- **Coherent** oscillations of AF vector ⇒ manifestation of **entanglement**?

Scientific goal

Ultrafast manipulation of the magnetic order



Science

Technology

Thermodynamics
Adiabatic approximation
Equilibrium

Terra incognita
Classical
?
Quantum

MRAM

Hard Drive

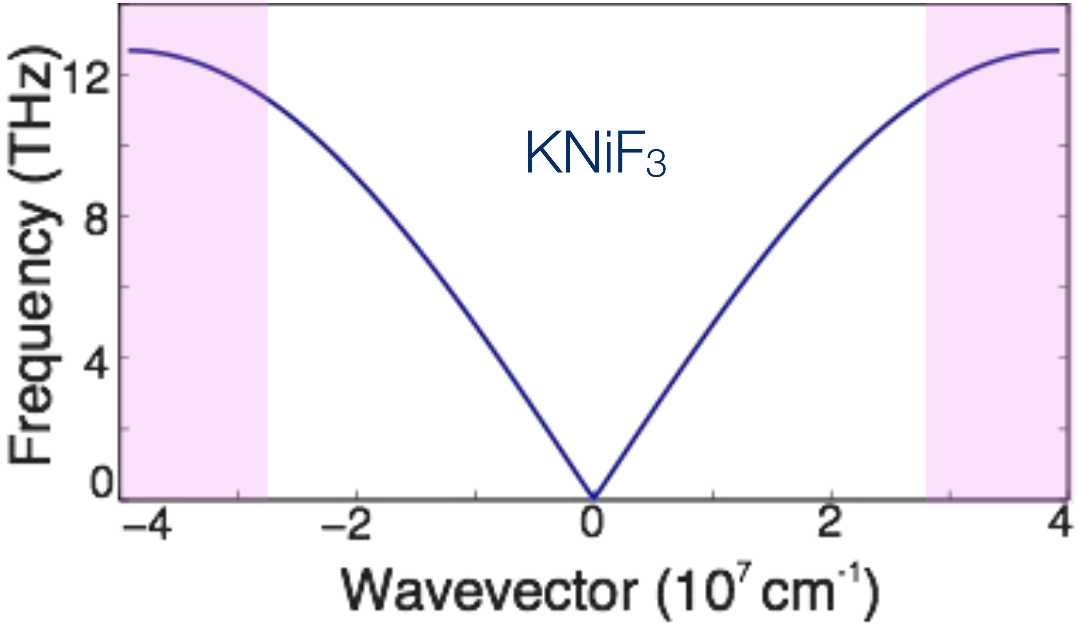
Computing time per bit

Recording time per bit

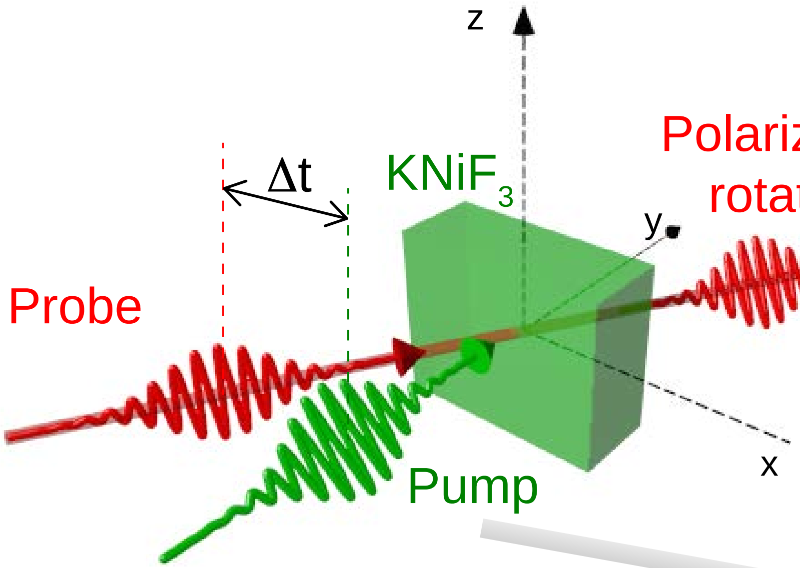
Challenge
Manipulate magnetism
fast
reproducible
low dissipation
quantum

Femto-nanomagnons in AF

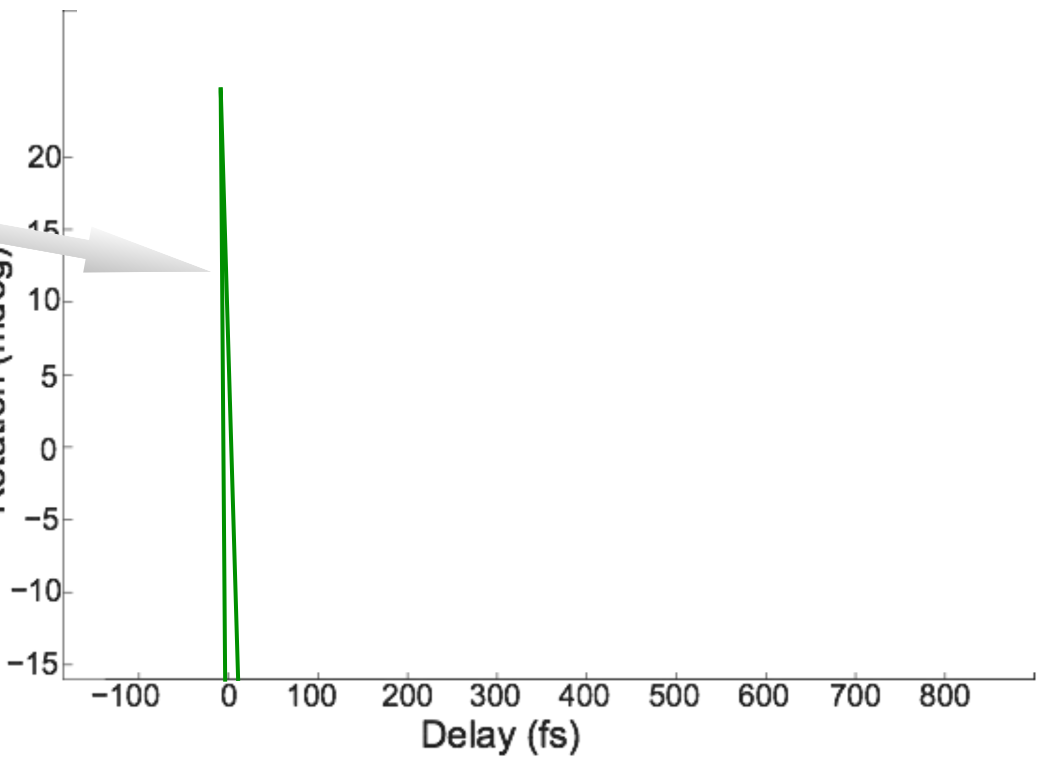
$$\omega_m \approx E_{ex}/\hbar$$

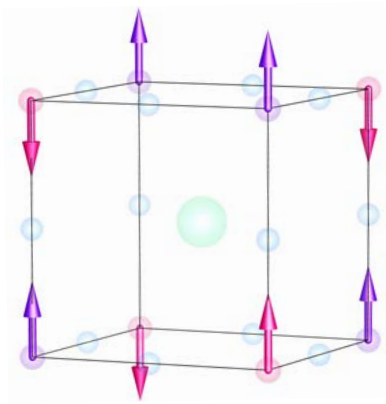


Experiment, pump-probe



$$\epsilon_{zz} = \epsilon_{zz}^{(0)} + \boxed{\lambda\langle S_{\uparrow}^{z} S_{\downarrow}^{z}\rangle}$$

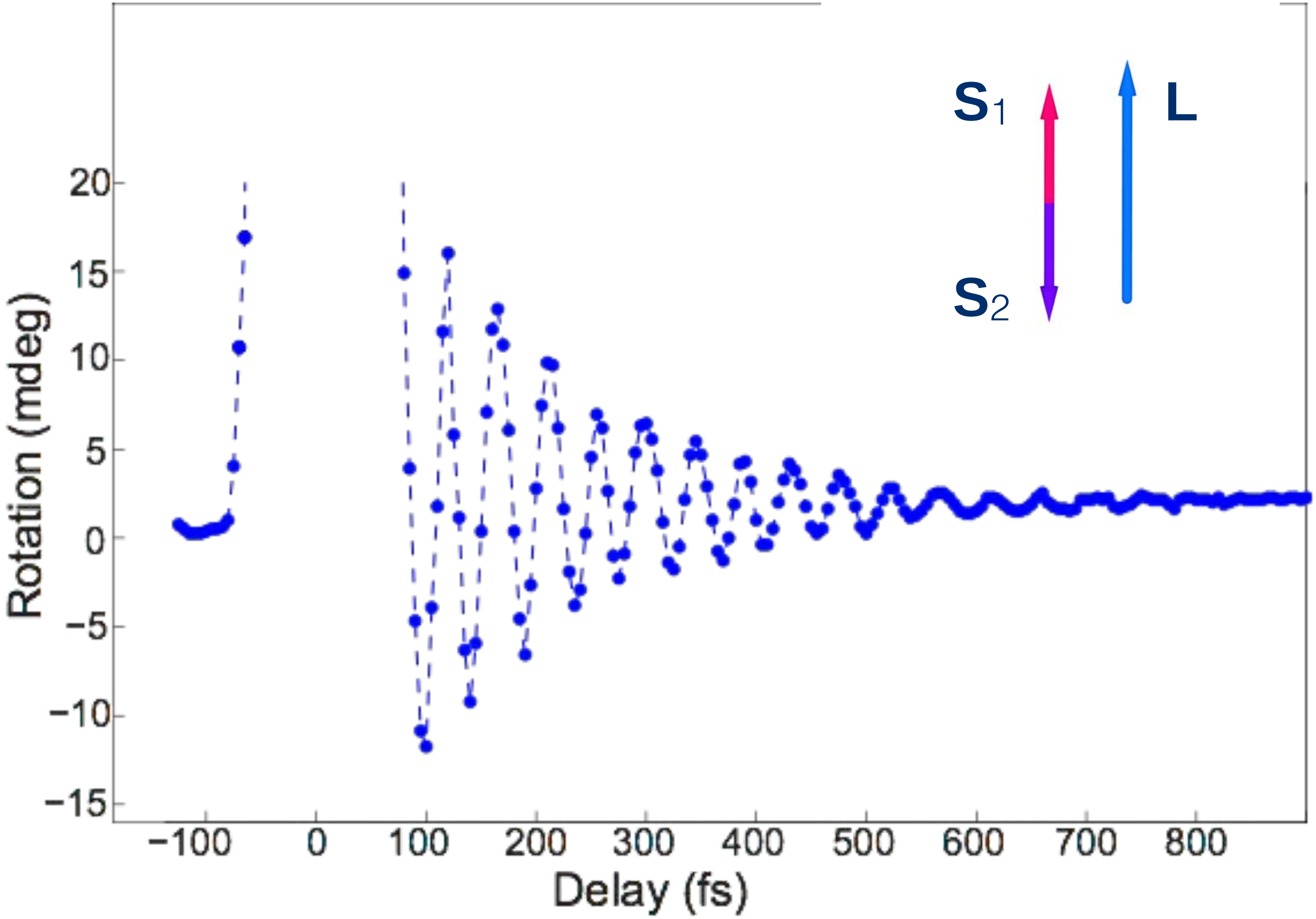




D. Bossini et al. PRB **89** (R), 060405 (2014)

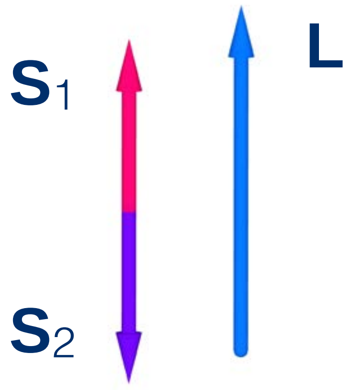
D. Bossini et al. Nat. Comm. **7,** *10645* (2016)

Experiment to be explained



Quantum or classical?

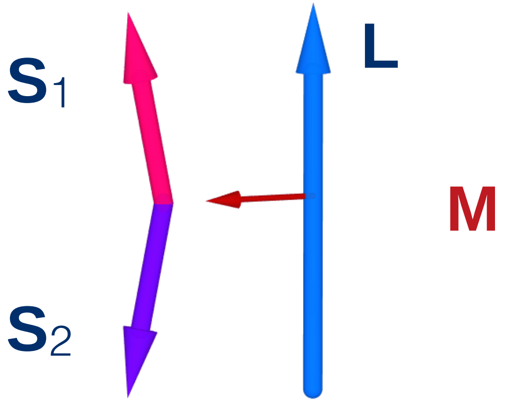
Quantum oscillations



$$\Delta L_z \propto \cos 2\Omega_{\mathbf{k}} t$$

$$\boxed{\mathbf{M}(t) = 0}$$

Classical oscillations



$$\Delta L_z \propto \cos 2\Omega_{\mathbf{k}} t$$

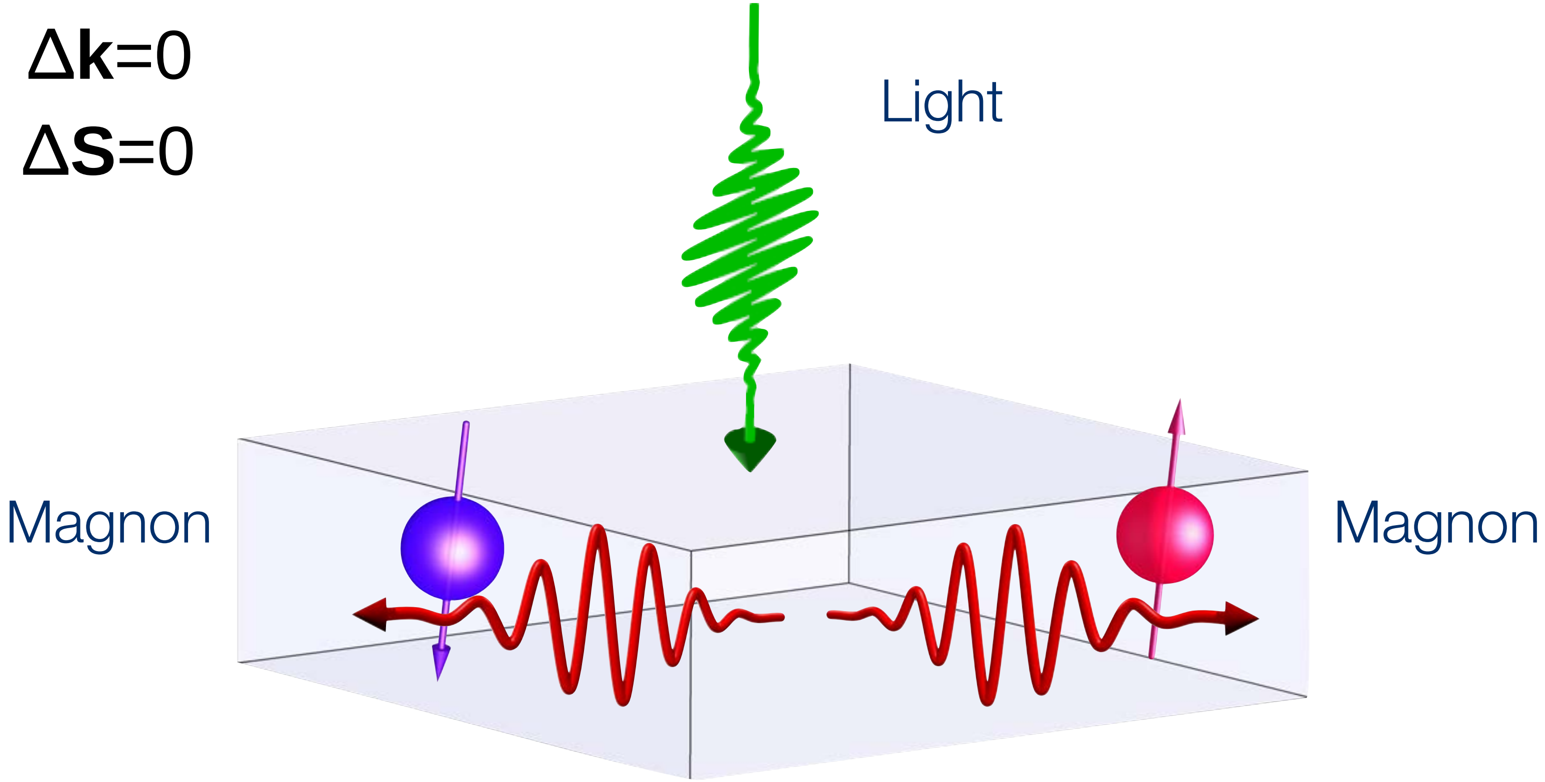
$$\boxed{\Delta M_x \propto \sin 2\Omega_{\mathbf{k}} t}$$

$$\xcancel{\mathbf{L}\times\ddot{\mathbf{L}} = -\gamma^2 H_{\mathrm{ex}}\mathbf{H}_L \times \mathbf{L}}$$

Two-magnon excitation

Δk=0

ΔS=0

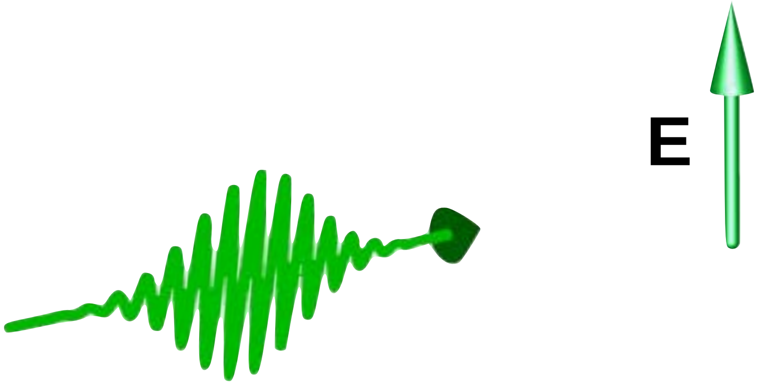


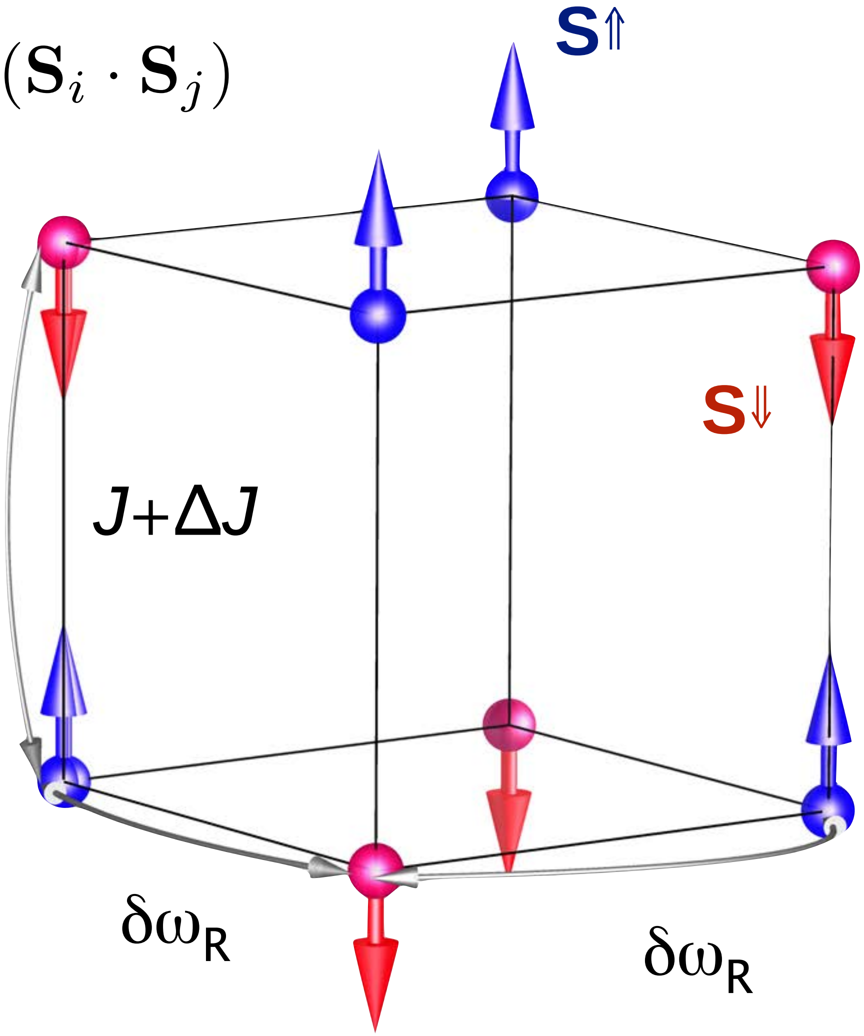
$$\delta\hat{H} = \boxed{\delta(t)}\Delta J \sum_{\langle i,j \rangle} (\hat{e}\cdot\hat{r}_{ij})^2 (\mathbf{S}_i \cdot \mathbf{S}_j)$$

P.A. Fleury and R. Loudon, Phys. Rev. 166, 514 (1968)
D. J. Lockwood and M.G. Cottam, Phys. Rev. B 35, 1973 (1987)

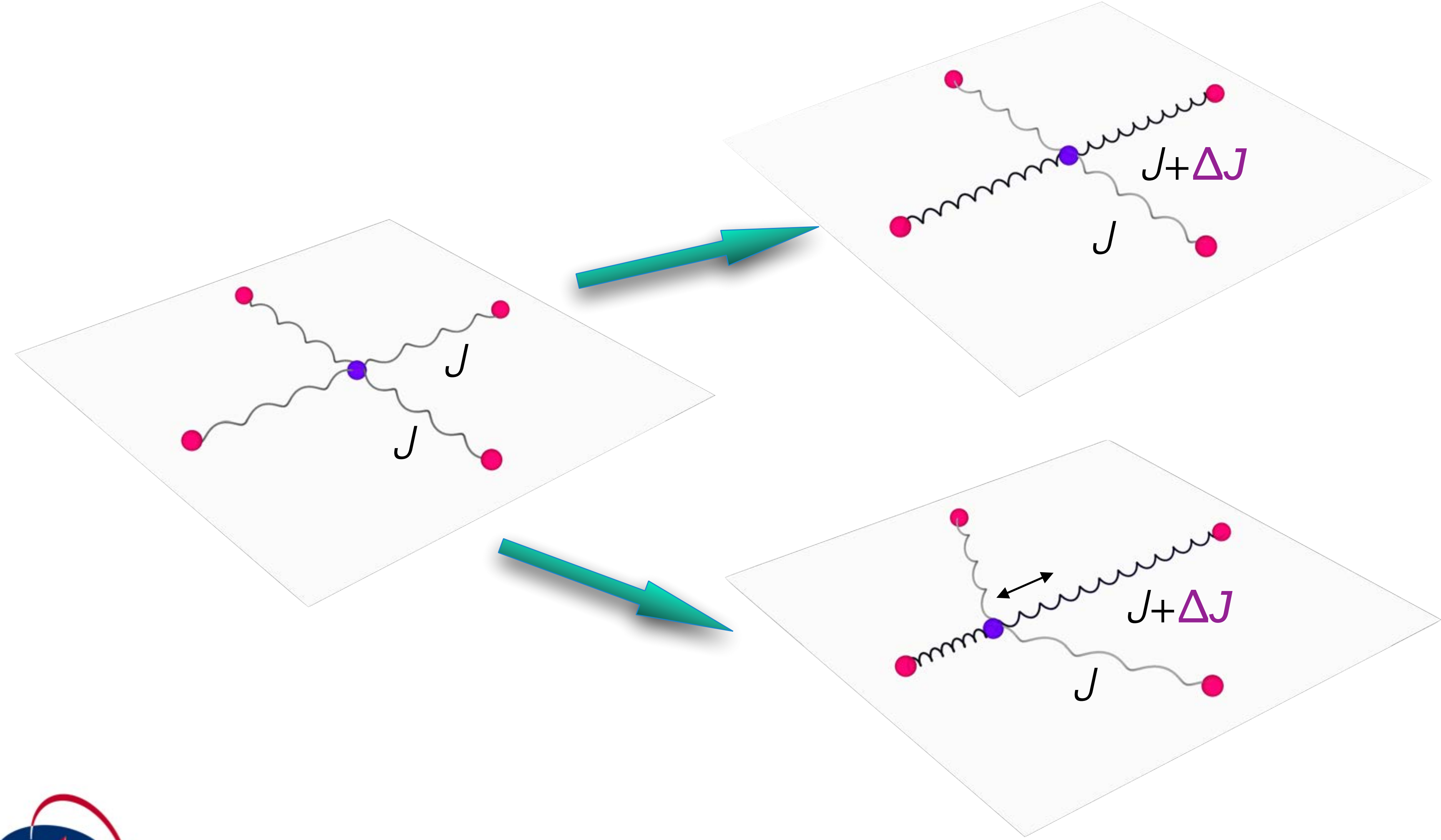
Light-matter interaction

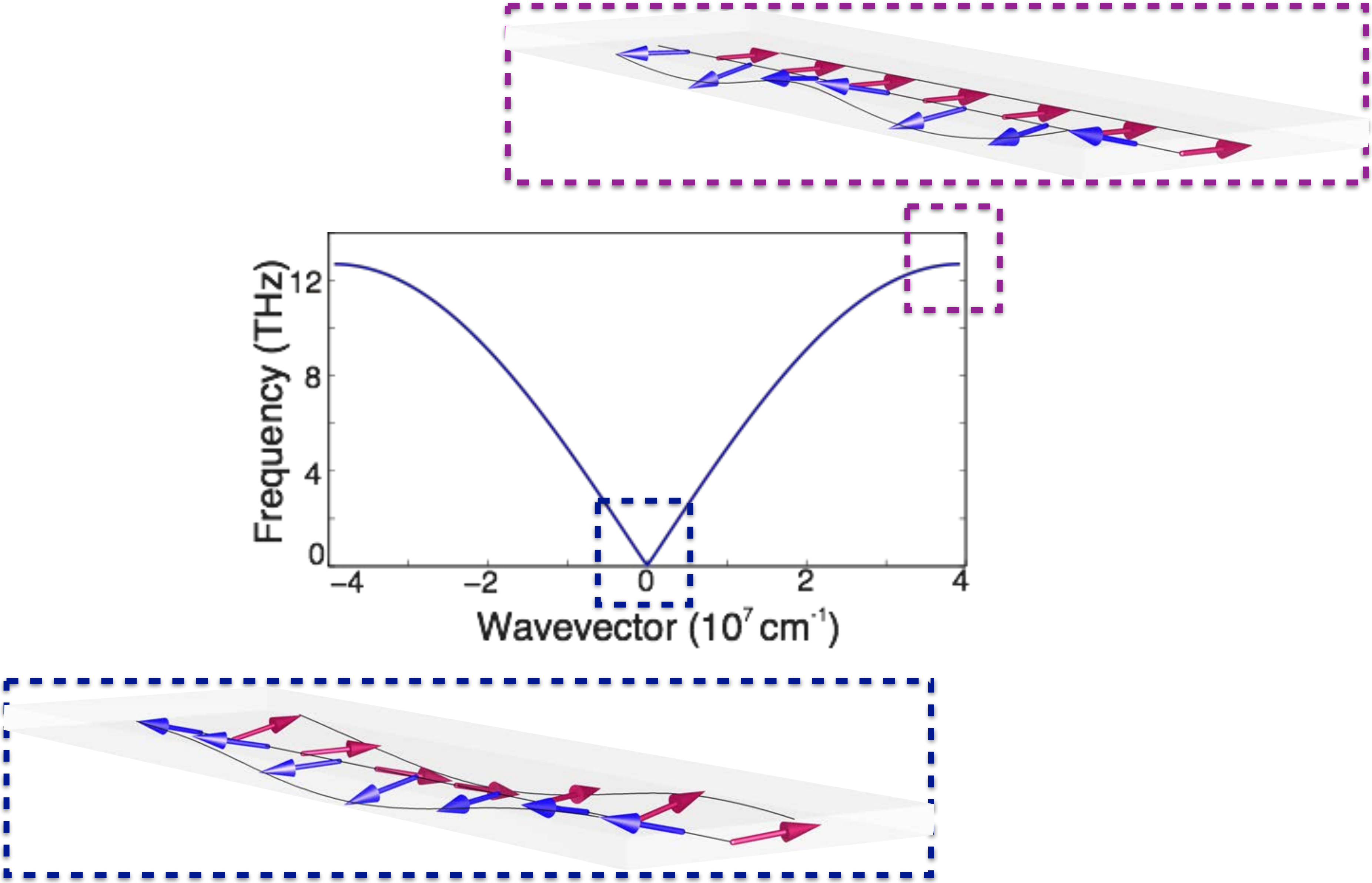
$$\delta\hat{H} = \delta(t)\Delta J \sum_{\langle i,j\rangle} (\hat{e}\cdot\hat{r}_{ij})^2(\mathbf{S}_i\cdot\mathbf{S}_j)$$



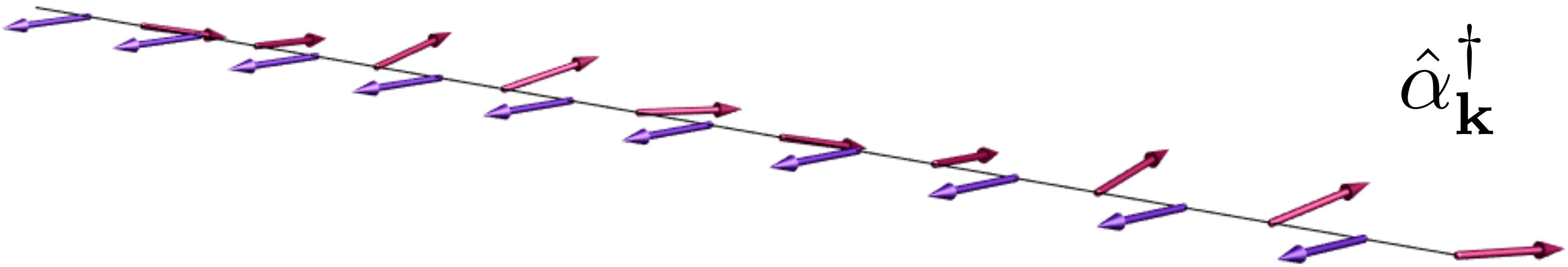


Change of exchange

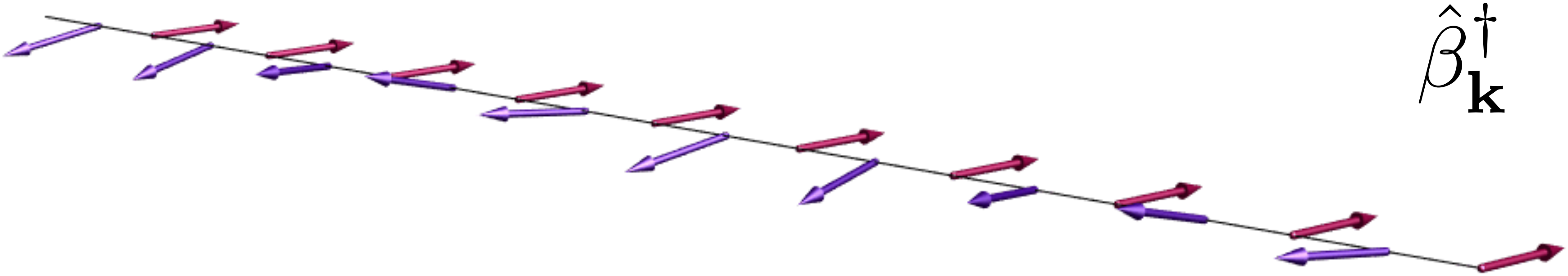



Frequency (THz)
12
8
4
0
−4
−2
0
2
4
Wavevector (10⁷ cm⁻¹)

Different magnons modes



$\hat{\alpha}^\dagger_{\mathbf{k}}$

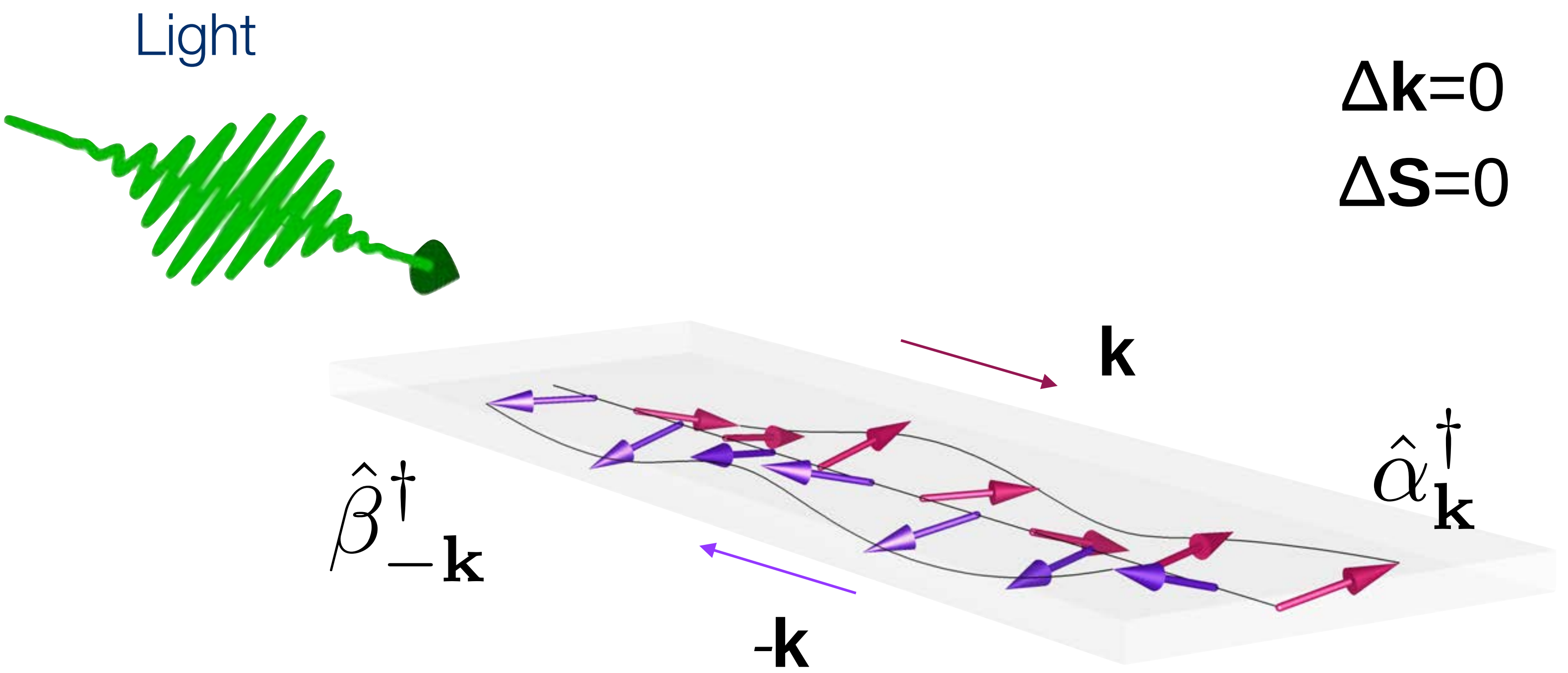


$\hat{\beta}^\dagger_{\mathbf{k}}$

$$\hat{H}_0 = 2\hbar \sum_{\mathbf{k}} \Omega_{\mathbf{k}} \left(\hat{\alpha}^\dagger_{\mathbf{k}} \hat{\alpha}_{\mathbf{k}} + \hat{\beta}^\dagger_{\mathbf{k}} \hat{\beta}_{\mathbf{k}} \right)$$

Excitation Hamiltonian

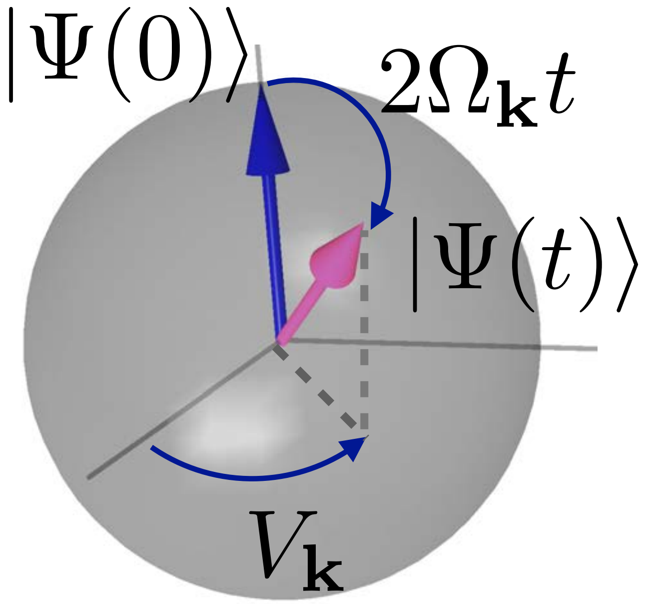
$$\delta\hat{H} = \delta(t)\sum_{\mathbf{k}}\left(V_{\mathbf{k}}\hat{\alpha}_{\mathbf{k}}\hat{\beta}_{-\mathbf{k}} + V^*_{\mathbf{k}}\hat{\alpha}^\dagger_{\mathbf{k}}\hat{\beta}^\dagger_{-\mathbf{k}}\right)$$



Δk=0

ΔS=0

Rotation in Hilbert space



Wave function

$$\boxed{|\Psi(t)\rangle = e^{-i2\Omega_{\mathbf{k}}t\hat{K}_z}e^{iV_{\mathbf{k}}\hat{K}^+}|\Psi(0)\rangle}$$

Hamiltonian

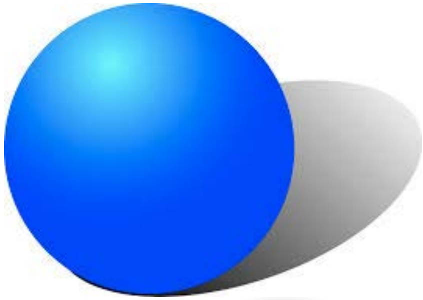
$$\hat{H}_0 = 2\hbar\sum_{\mathbf{k}}\Omega_{\mathbf{k}}\underbrace{(\alpha^\dagger_{\mathbf{k}}\alpha_{\mathbf{k}} + \beta^\dagger_{\mathbf{k}}\beta_{\mathbf{k}})}_{\hat{K}_z}$$

Excitation

$$\delta\hat{H} = \delta(t)\sum_{\mathbf{k}}(V_{\mathbf{k}}\underbrace{\alpha_{\mathbf{k}}\beta_{-\mathbf{k}}}_{\hat{K}^+} + V^*_{\mathbf{k}}\underbrace{\alpha^\dagger_{\mathbf{k}}\beta^\dagger_{-\mathbf{k}}}_{\hat{K}^-})$$

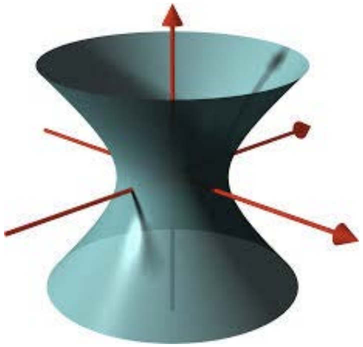
Perelomov's states: SU(2) vs SU(1,1)

SU(2)



$$[\hat{S}^z, \hat{S}^\pm] = \pm \hat{S}^\pm, \quad [\hat{S}^-, \hat{S}^+] = \boxed{-2}\hat{S}^z$$

SU(1,1)



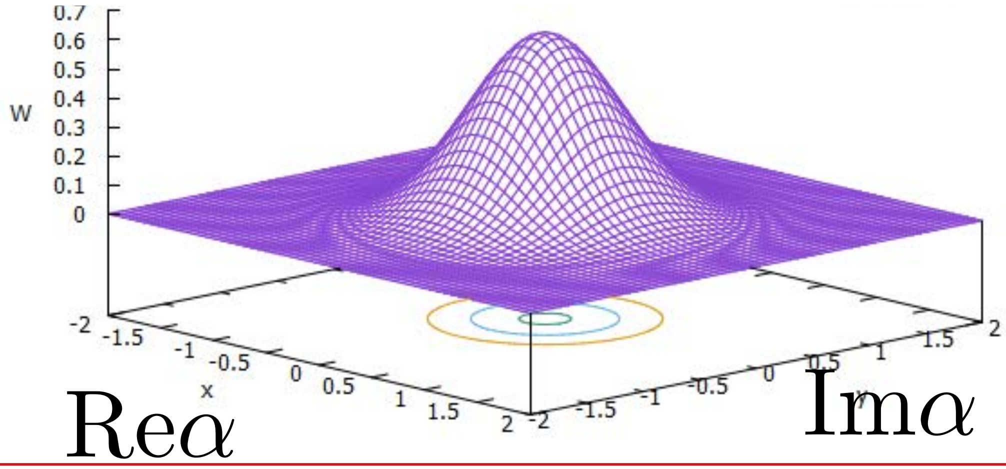
$$[\hat{K}^z_{\mathbf{k}}, \hat{K}^\pm_{\mathbf{k}}] = \pm \hat{K}^\pm_{\mathbf{k}}, \quad [\hat{K}^-_{\mathbf{k}}, \hat{K}^+_{\mathbf{k}}] = \boxed{2}\hat{K}^z_{\mathbf{k}}.$$

Coherent state

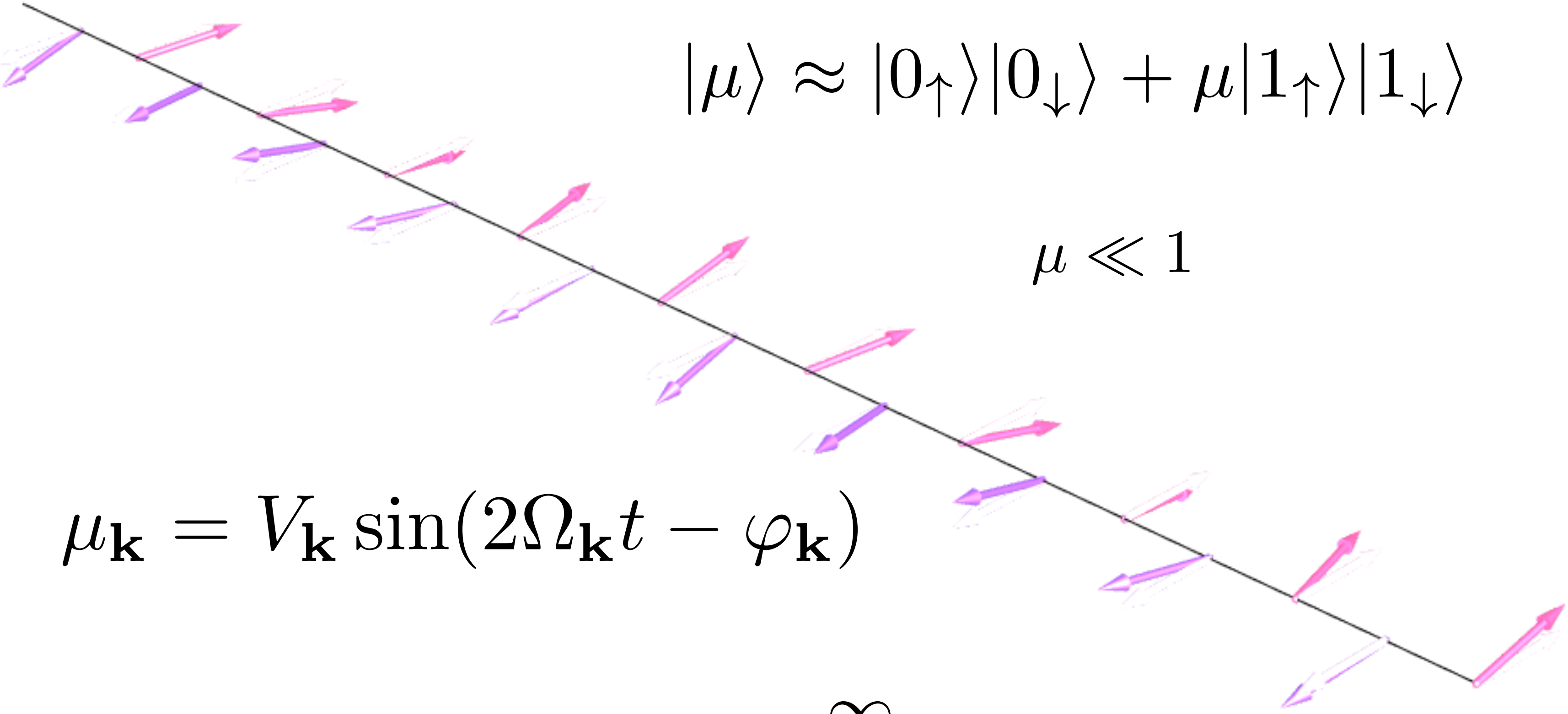
Photons, coherent state

$$|\alpha\rangle = \exp[\alpha\hat{a}^{\dagger} - \alpha^{*}\hat{a}]|0\rangle$$

$$|\alpha\rangle = e^{-|\alpha|^2/2}\sum_{n=0}^{\infty}\frac{\alpha^n}{\sqrt{n!}}|n\rangle$$



Coherent state and entangled magnons



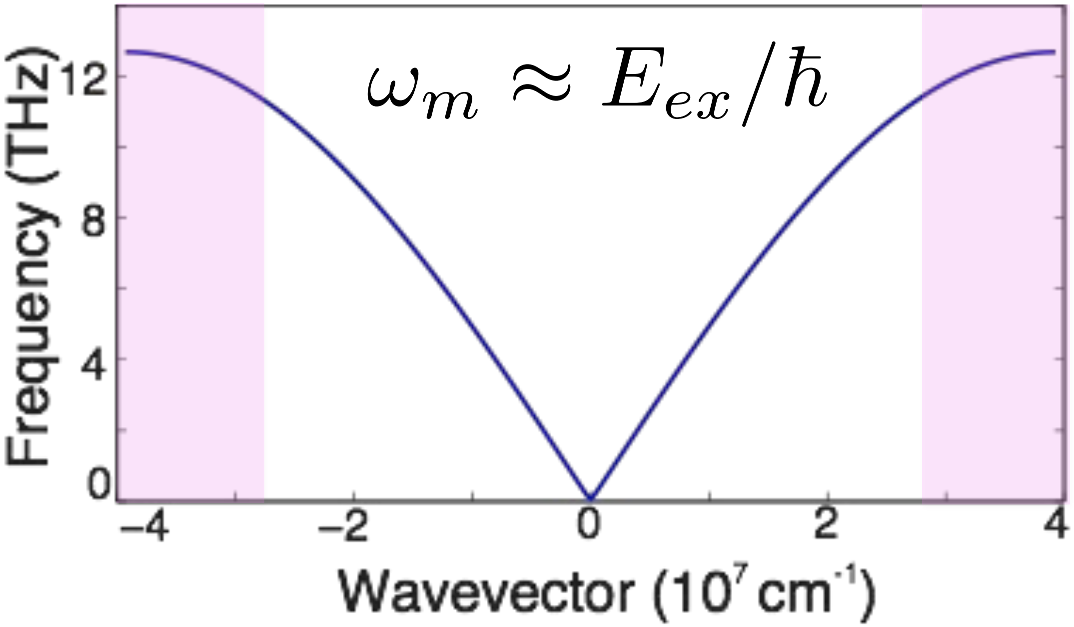
$$|\mu\rangle \approx |0_\uparrow\rangle|0_\downarrow\rangle + \mu|1_\uparrow\rangle|1_\downarrow\rangle$$

$$\mu \ll 1$$

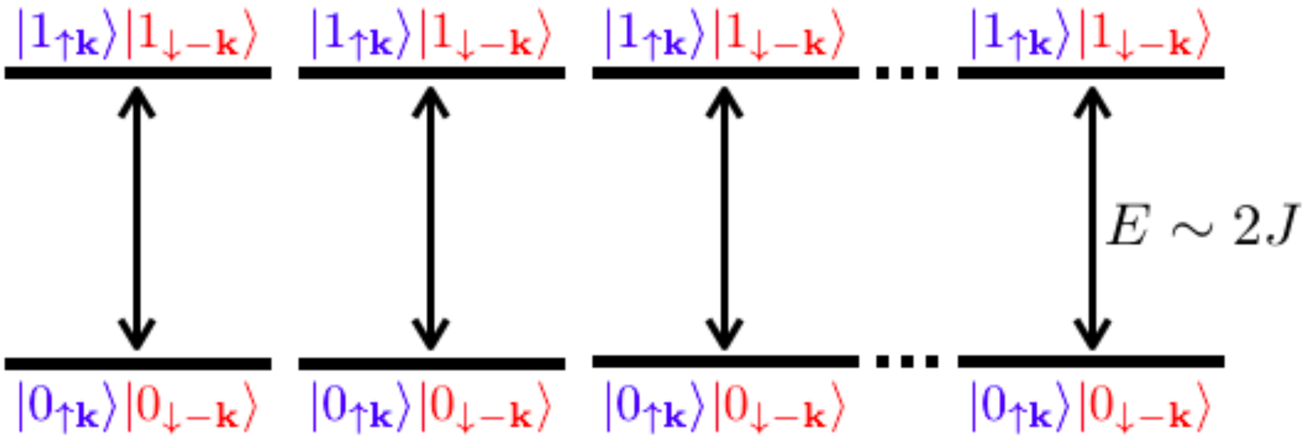
$$\mu_{\mathbf{k}} = V_{\mathbf{k}} \sin(2\Omega_{\mathbf{k}} t - \varphi_{\mathbf{k}})$$

$$|\mu\rangle = \sqrt{1-|\mu|^2}\sum_{n=0}^{\infty} \mu^n |n_\uparrow\rangle|n_\downarrow\rangle$$

Edge of BZ, phase matching

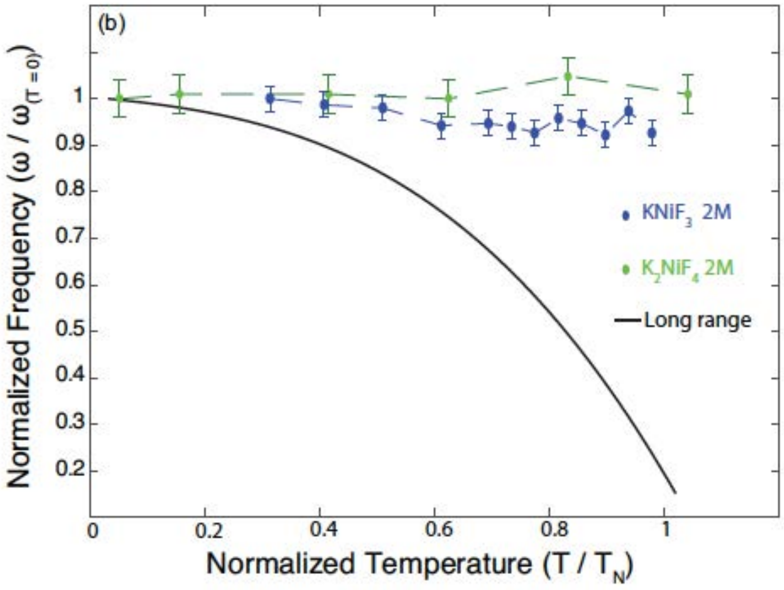


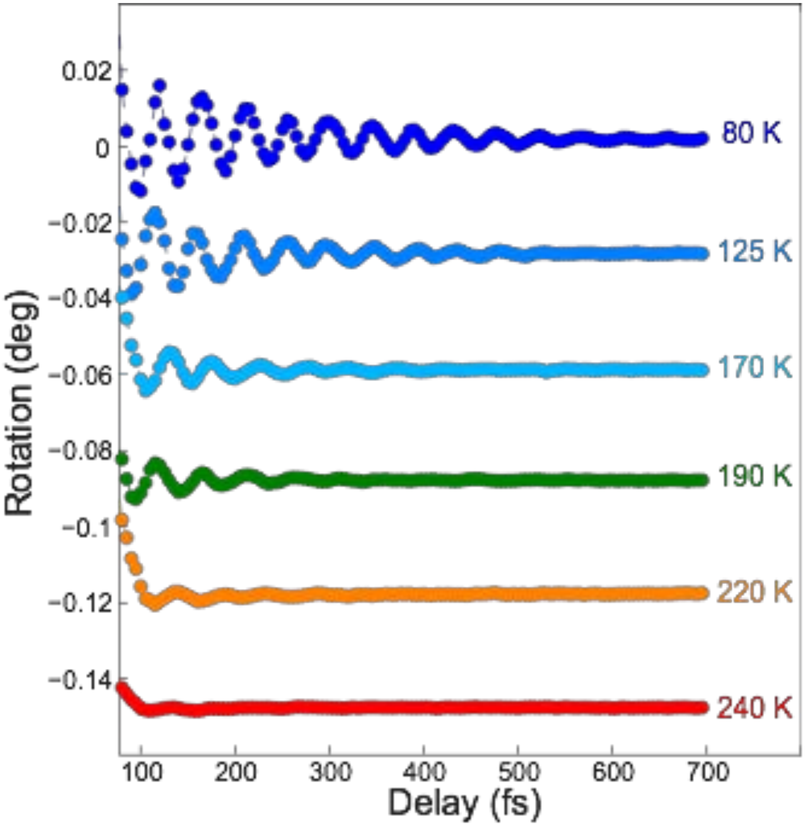
Frequency: **22 THz**
Wavelength: **1 nm**



Temperature dependence

Frequency: 22 THz, wavelength: 1 nm





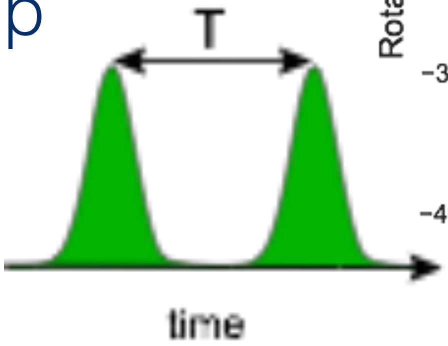
Control the dynamics

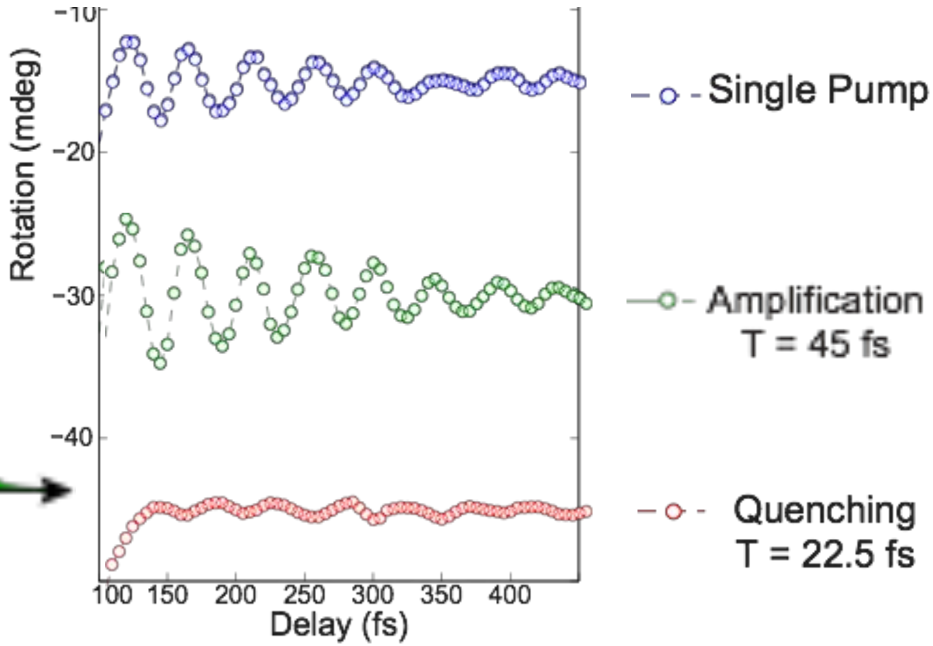
$$i\hbar\partial_t\mu = 2\hbar\left(\Omega_{\mathbf{k}} + \delta\Omega_{\mathbf{k}} f(t)\right)\mu + \hbar V f(t)(1+\mu^2)$$

$$\mu \to \mu + \mu e^{-i2\Omega_{\mathbf{k}} T}$$

$$\Delta L_z(t) = -V_{\mathbf{k}} \cos^2 \Omega_{\mathbf{k}} T \sin 2\Omega_{\mathbf{k}} t$$

Two pump pulses





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Coherent vs entangled

Photons, coherent state

$$|\alpha\rangle = e^{-|\alpha|^2/2} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle$$

Photons, coherent entangled state

$$|\beta\rangle = e^{-|\beta|^2/2} \sum_{n=0}^{\infty} \frac{\beta^n}{\sqrt{n!}} |n\rangle|n\rangle$$

Magnons, coherent entangled state

$$|\mu\rangle = \sqrt{1-|\mu|^2} \sum_{n=0}^{\infty} \mu^n |n_\uparrow\rangle|n_\downarrow\rangle$$

Coherent photons vs magnons

Photons

- Minimal uncertainty

$$\langle\Delta\hat{p}\rangle\langle\Delta\hat{x}\rangle = \hbar/2$$

- Eigen state of annihilation operator

$$\hat{a}|\alpha\rangle = \alpha|\alpha\rangle$$

- Generated by the displacement operator

$$\hat{D}(\alpha')|\alpha\rangle = |\alpha + \alpha'\rangle$$

Magnons

- ~~Minimal uncertainty~~

$$\langle\Delta\hat{K}^{+}\rangle\langle\Delta\hat{K}^{-}\rangle = f(\mu)\hbar/2$$

- Eigen state of annihilation operator

$$\hat{K}^{-}|\mu\rangle \propto \mu|\mu\rangle$$

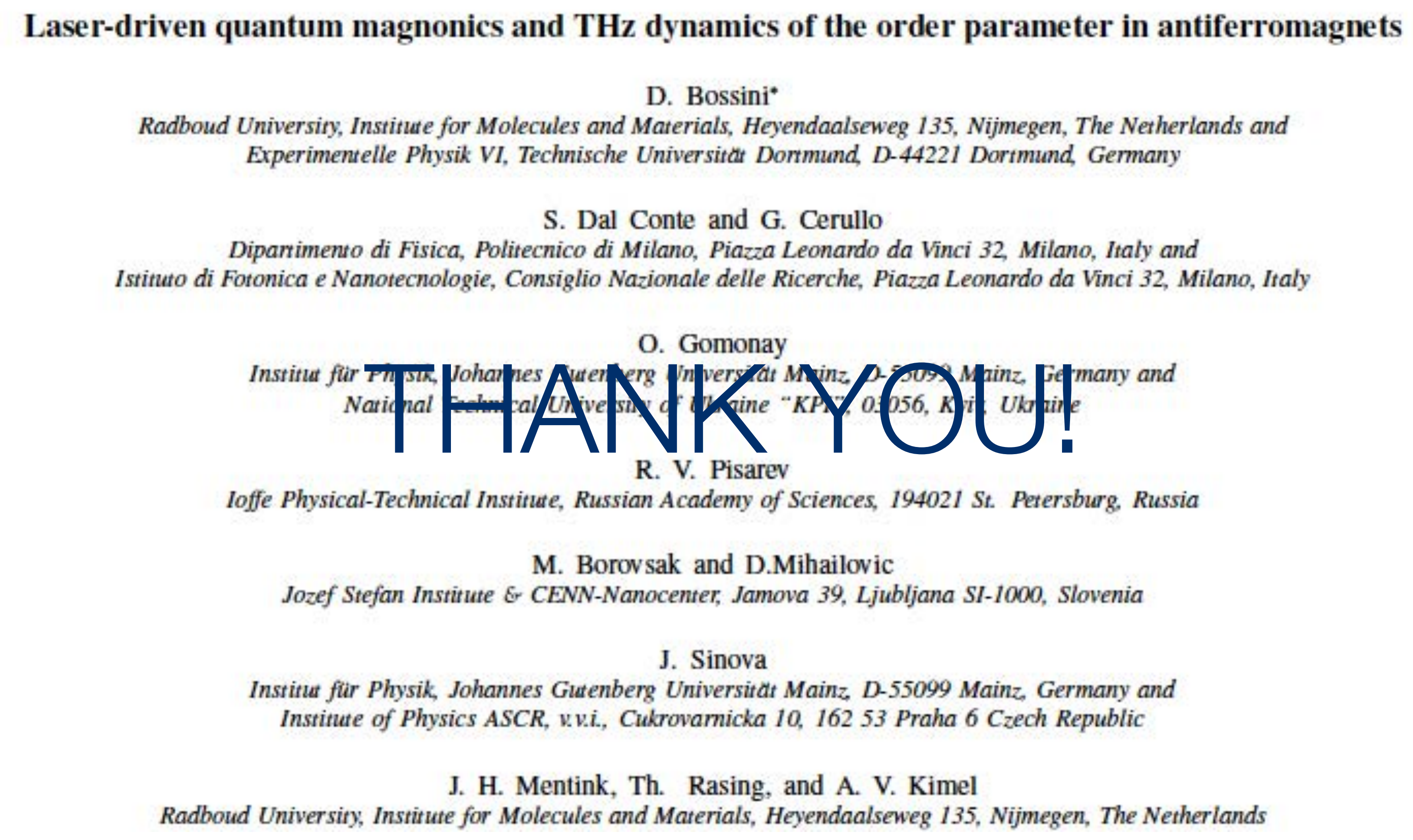
- Generated by the displacement operator?

$$\hat{D}(\mu)|0\rangle = |\mu\rangle$$

Conclusions

- Light => two-magnon excitations
- Macroscopic oscillations => **quantum** effect
- Macroscopic description with **coherent** states
- **New entangled** state in solids?

Collaboration



Laser-driven quantum magnonics and THz dynamics of the order parameter in antiferromagnets

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THANK YOU!

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THANK YOU

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Femtosecond quantum spin dynamics induced by femto-nanomagnons in Heisenberg antiferromagnets

I. Intro to talk

II. Raman excitation of 2M modes

III. Spin waves and two-magnon excitations

IV. Coherent state and macroscopic description

V. Conclusion