Theoretical description of magnetic precessions during ultrafast laser excitation

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Ultrafast inverse Faraday effect

Ultrafast light-induced precessions via inverse Faraday effect

$$\mathbf{M}(\mathbf{H}=0) = \frac{\mathbf{B}}{4\pi} = \frac{i\epsilon_{03}}{16\pi}\mathbf{E}_0^* \times \mathbf{E}_0$$

How can magnetization be nonzero, after the action of light, when $E_0(t) = 0$?

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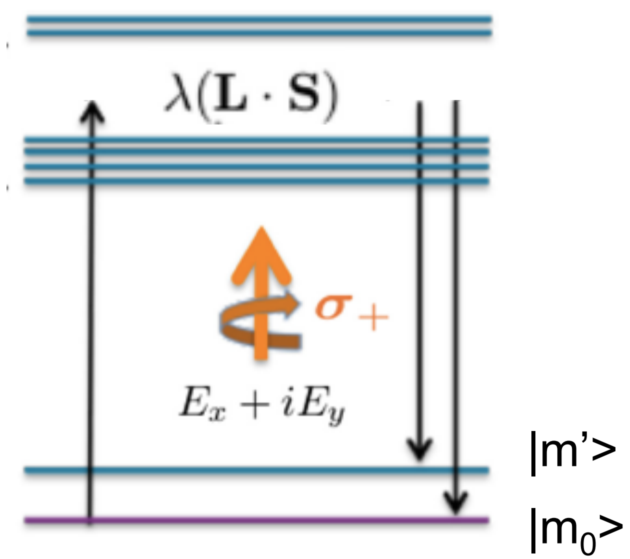
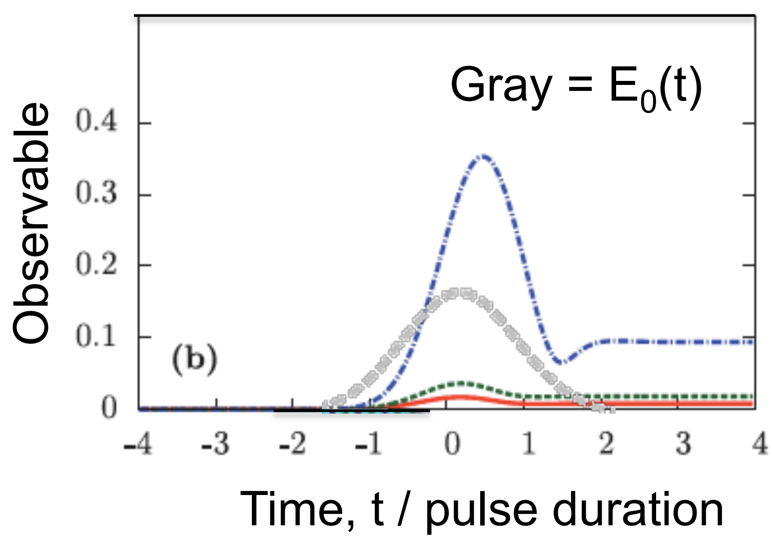
Stimulated Raman scattering process



Gaussian-shaped pump pulse



D. Popova (=D. Gorelova), A. Bringer, and S. Blügel, Phys. Rev. B 84, 214421 (2011).
D. Popova, A. Bringer, and S. Blügel, Phys. Rev. B 85, 094419 (2012).

Ultrafast inverse Faraday effect

Ultrafast light-induced precessions via inverse Faraday effect

$$\mathbf{M}(\mathbf{H}=0) = \frac{\mathbf{B}}{4\pi} \neq \frac{i\epsilon_{03}}{16\pi}\mathbf{E}_0^* \times \mathbf{E}_0$$

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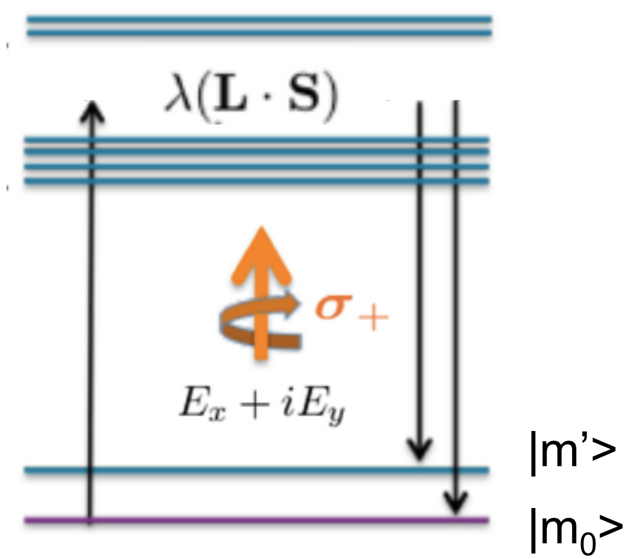

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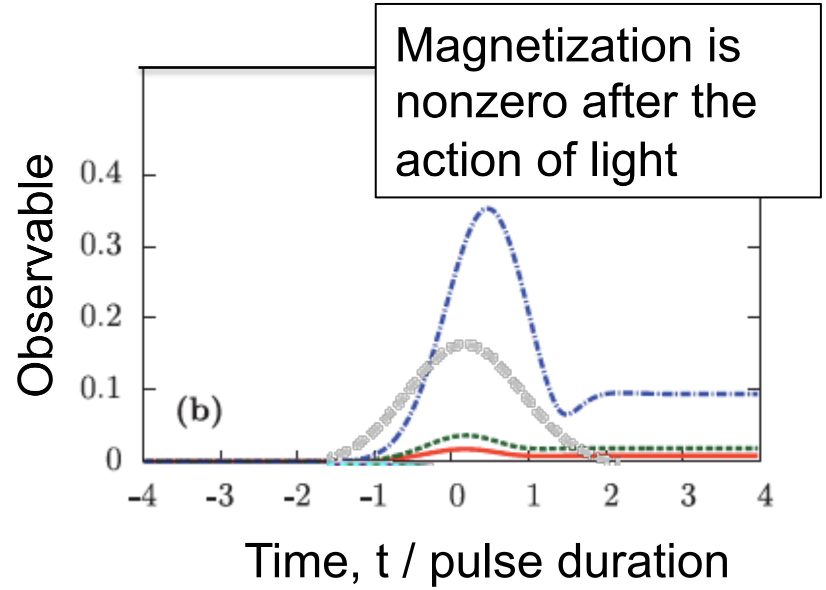

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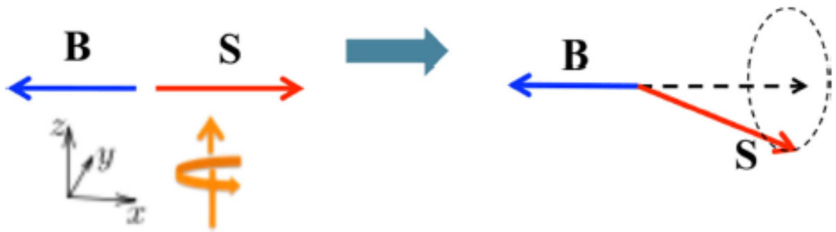
Single spin in an external magnetic field



Hamiltonian: spin-orbit coupling + Zeeman interaction $-\hat{\mathbf{d}}\cdot\mathbf{E}(t)$

D. Popova-Gorelova, A. Bringer, S. Blügel, Phys Rev B 104, 224418 (2021)

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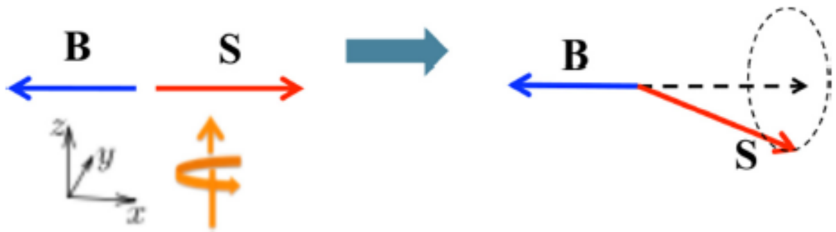


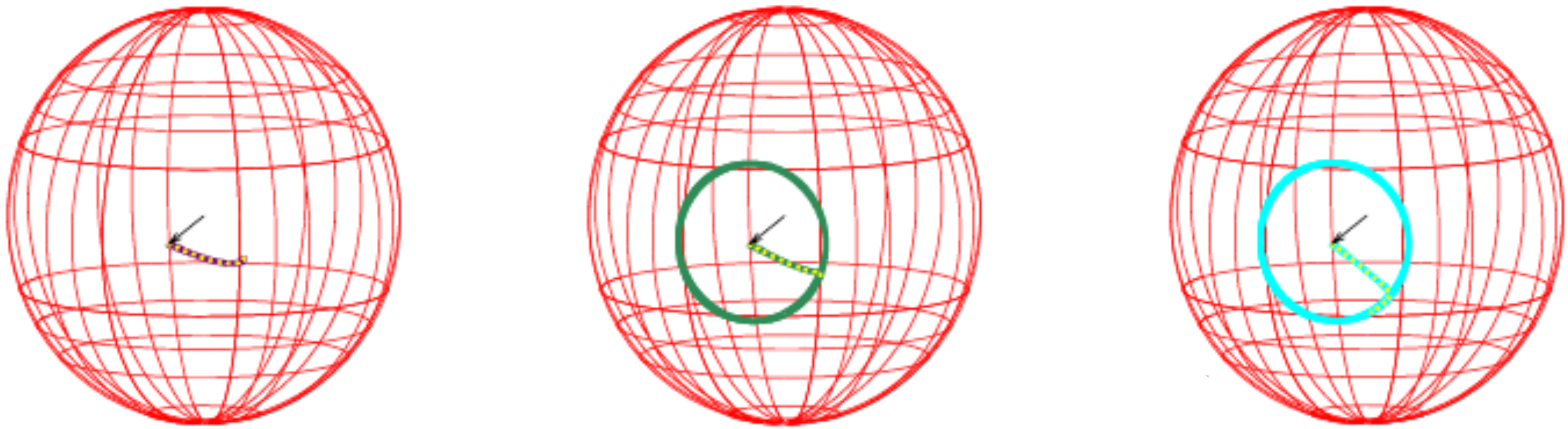
Pump pulse duration T_{dr} = 117 fs

T_L – period of Larmor precession

Only light

T_L / T_{dr} = 15

T_L / T_{dr} = 40



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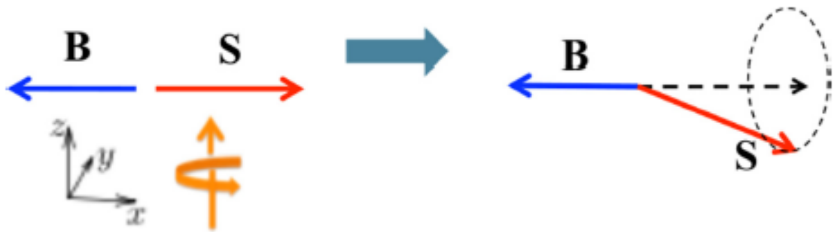
Oscillations during the action of the pump pulse



Pump pulse duration T_{dr} = 117 fs

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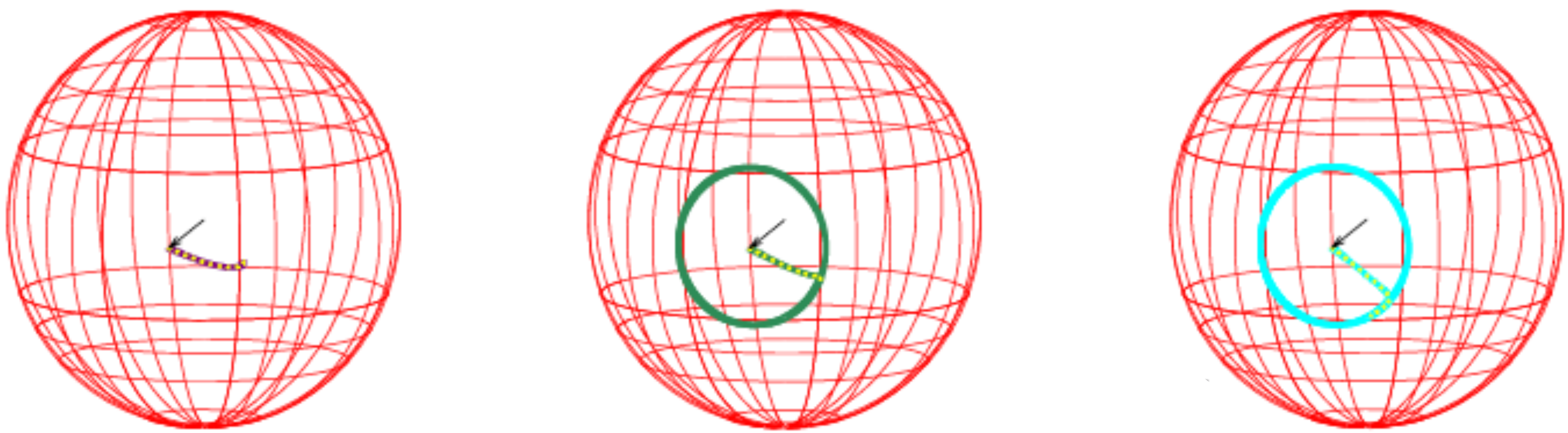
Spin starts to oscillate during the action of a pulse with a duration **40** times shorter

Only light | T_L / T_{dr} = 40 | T_L / T_{dr} = 15



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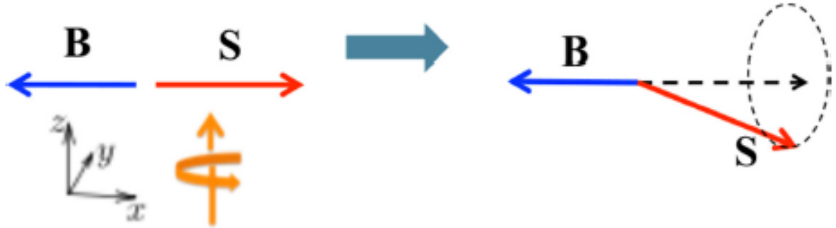

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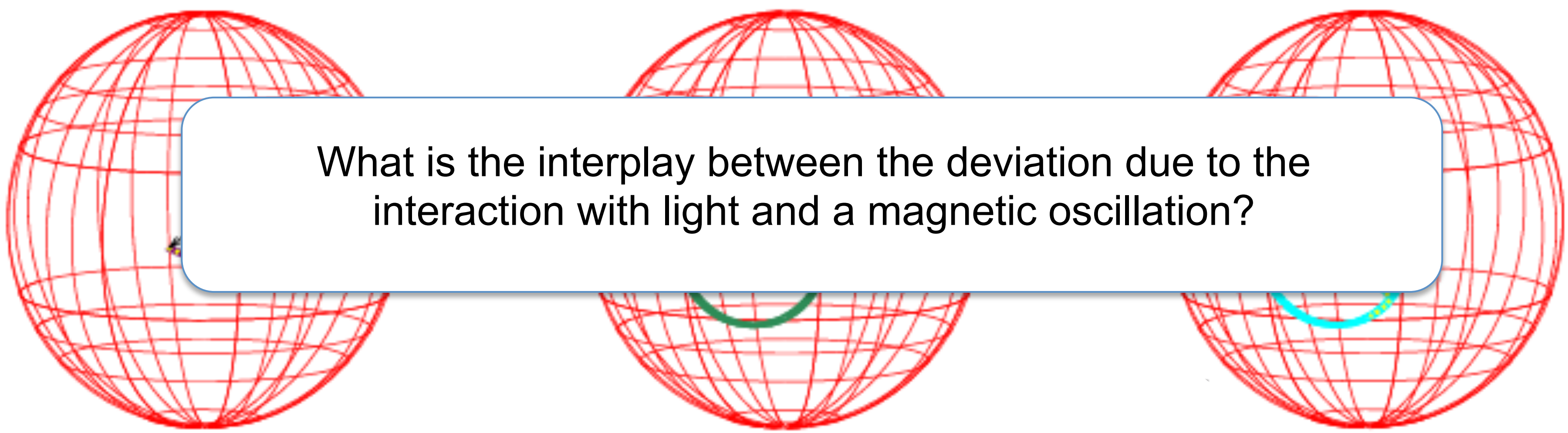

What is the interplay between the deviation due to the interaction with light and a magnetic oscillation?

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Time evolution of an observable

Heisenberg picture

$$\frac{d}{dt}\langle \hat{O} \rangle = i\,[\hat{H}, \hat{O}]$$

We are interested in the dynamics of a magnetic moment component

$$J_\alpha(t) = \langle \hat{O} \rangle$$

Substitute $\hat{H}$ for an effective magnetic Hamiltonian $\hat{H}_m$

$\hat{H}_m$ couples only to magnetic moments

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Time evolution of an observable

Heisenberg picture

$$\frac{dJ_\alpha(t)}{dt} = i\,[\hat{H}_m, \hat{J}_\alpha]$$

Advantages:

Equation of motion for magnetic vectors

Application of effective magnetic Hamiltonians

Taking into account dissipation

Problem:

Time-dependent electric field of light couples to electrons $-\hat{\mathbf{d}} \cdot \mathbf{E}(t)$

Idea:

Derive a time-dependent effective magnetic Hamiltonian
that describes the action of light on a magnetic system

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Time evolution of an observable

$$\langle \hat{O} \rangle = \langle \psi(t) | \hat{O} | \psi(t) \rangle$$

Schrödinger picture

$$\frac{d\psi(t)}{dt} = -i\hat{H}\psi(t)$$

Equivalent

Heisenberg picture

$$\frac{d}{dt}\langle \hat{O} \rangle = i\,[\hat{H}, \hat{O}]$$

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Derivation of an effective magnetic Hamiltonian

Time-dependent Schrödinger equation

Obtain the time evolution of electronic wave functions

Time evolution of magnetic vectors as expectation values

Derive a time-dependent effective magnetic Hamiltonian such that the solution of a Heisenberg picture coincides with the time-dependent Schrödinger equation

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Spin matrices

$$S_x = \frac{\hbar}{2}\begin{pmatrix} 0 & \sqrt{3} & 0 & 0 \\ \sqrt{3} & 0 & 2 & 0 \\ 0 & 2 & 0 & \sqrt{3} \\ 0 & 0 & \sqrt{3} & 0 \end{pmatrix},$$

Sz = +3/2
Sz = +1/2
Sz = -1/2
Sz = -3/2

$$S_y = \frac{\hbar}{2}\begin{pmatrix} 0 & -i\sqrt{3} & 0 & 0 \\ i\sqrt{3} & 0 & -2i & 0 \\ 0 & 2i & 0 & -i\sqrt{3} \\ 0 & 0 & i\sqrt{3} & 0 \end{pmatrix}$$

$$S_z = \frac{\hbar}{2}\begin{pmatrix} 3 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -3 \end{pmatrix},$$

Effective magnetic Hamiltonian

General solution represented in the Hilbert space of spin/momentum projections

$$\hat{\mathcal{H}}_J = -\sum_a^n \gamma_a \hat{N}_a + \frac{1}{2}\sum_{a,b}^n (\nu_a - \nu_b)(\langle \hat{N}_{ab-}\rangle \hat{N}_{ab+} - \langle \hat{N}_{ab+}\rangle \hat{N}_{ab-})$$

For total spin 3/2

$$\hat{N}_{12+} = \begin{pmatrix} 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix},$$

$$\hat{N}_{12-} = \begin{pmatrix} 0 & -i & 0 & 0 \\ i & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix},$$

$$\hat{N}_1 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}.$$

$$\hat{S}_x = \frac{\sqrt{3}}{2}\hat{N}_{12+} + \hat{N}_{23+} + \frac{\sqrt{3}}{2}\hat{N}_{34+}$$

$$\hat{S}_y = \frac{\sqrt{3}}{2}\hat{N}_{12-} + \hat{N}_{23-} + \frac{\sqrt{3}}{2}\hat{N}_{34-}$$

$$\hat{S}_z = \frac{3}{2}\hat{N}_1 + \frac{1}{2}\hat{N}_2 - \frac{1}{2}\hat{N}_3 - \frac{3}{2}\hat{N}_4$$

D. Popova-Gorelova, A. Bringer, S. Blügel, Phys Rev B 104, 224418 (2021)

Effective magnetic Hamiltonian

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Solution in the case of a single spin

$$\hat{\mathcal{H}}_J = f(t)(S_y\hat{S}_x - S_x\hat{S}_y) + g(t)\hat{S}_z + h(t)\hat{S}^2$$

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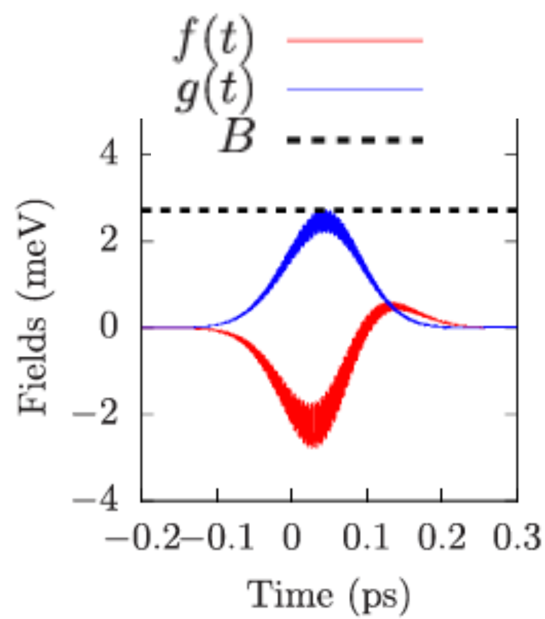
Effective magnetic Hamiltonian

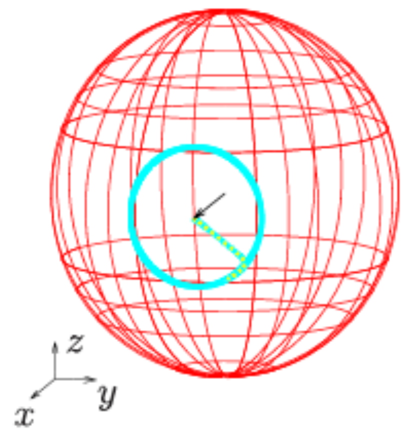
$$\hat{\mathcal{H}}_J = f(t)(S_y\hat{S}_x - S_x\hat{S}_y) + g(t)\hat{S}_z + h(t)\hat{S}^2$$

$$\frac{dJ_\alpha(t)}{dt} = i\,[\hat{H}_m, \hat{J}_\alpha]$$

Magnetic Hamiltonian represented via spin/angular momentum operators

$$S'_x = -f(t)S_xS_z - g(t)S_y,$$
$$S'_y = -f(t)S_yS_z + g(t)S_x + BS_z,$$
$$S'_z = -f(t)(S_x^2 + S_y^2) - BS_y.$$





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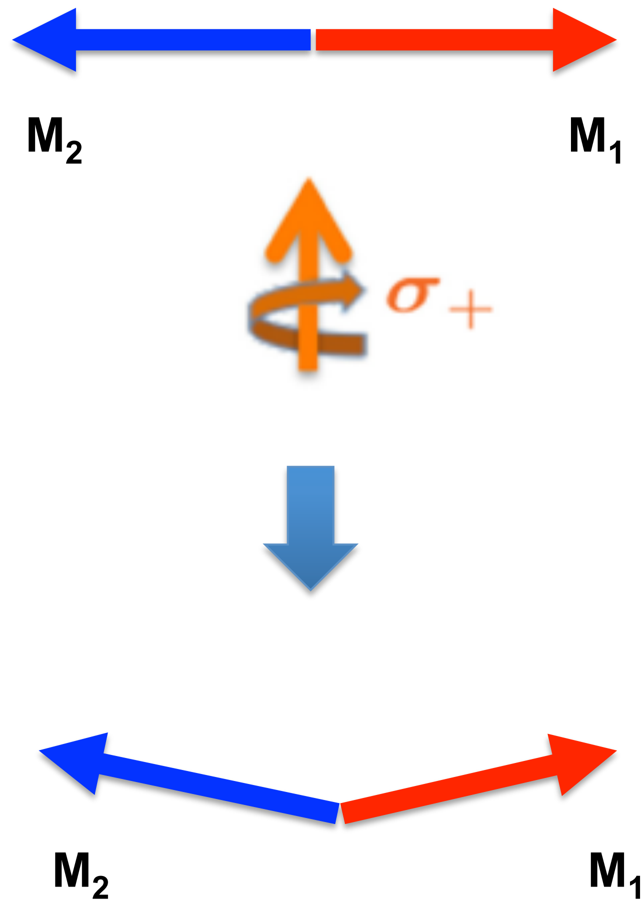
Two-sublattice antiferromagnet



Due to the symmetry of the effective operator, magnetic vectors deviate such that

$$M_{1x} = -M_{2x}$$
$$M_{1y} = -M_{2y}$$
$$M_{1z} = M_{2z}$$

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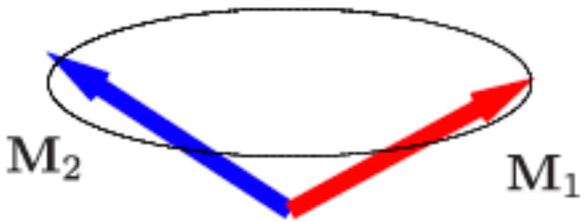
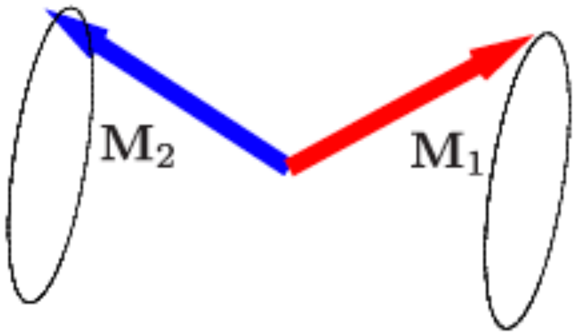
Precession modes antiferromagnet

Exchange interaction
(mean field approach)



Crystal field
(easy-axis
antiferromagnet)

$\Delta(3\hat{J}_x^2 - \hat{J}^2)$



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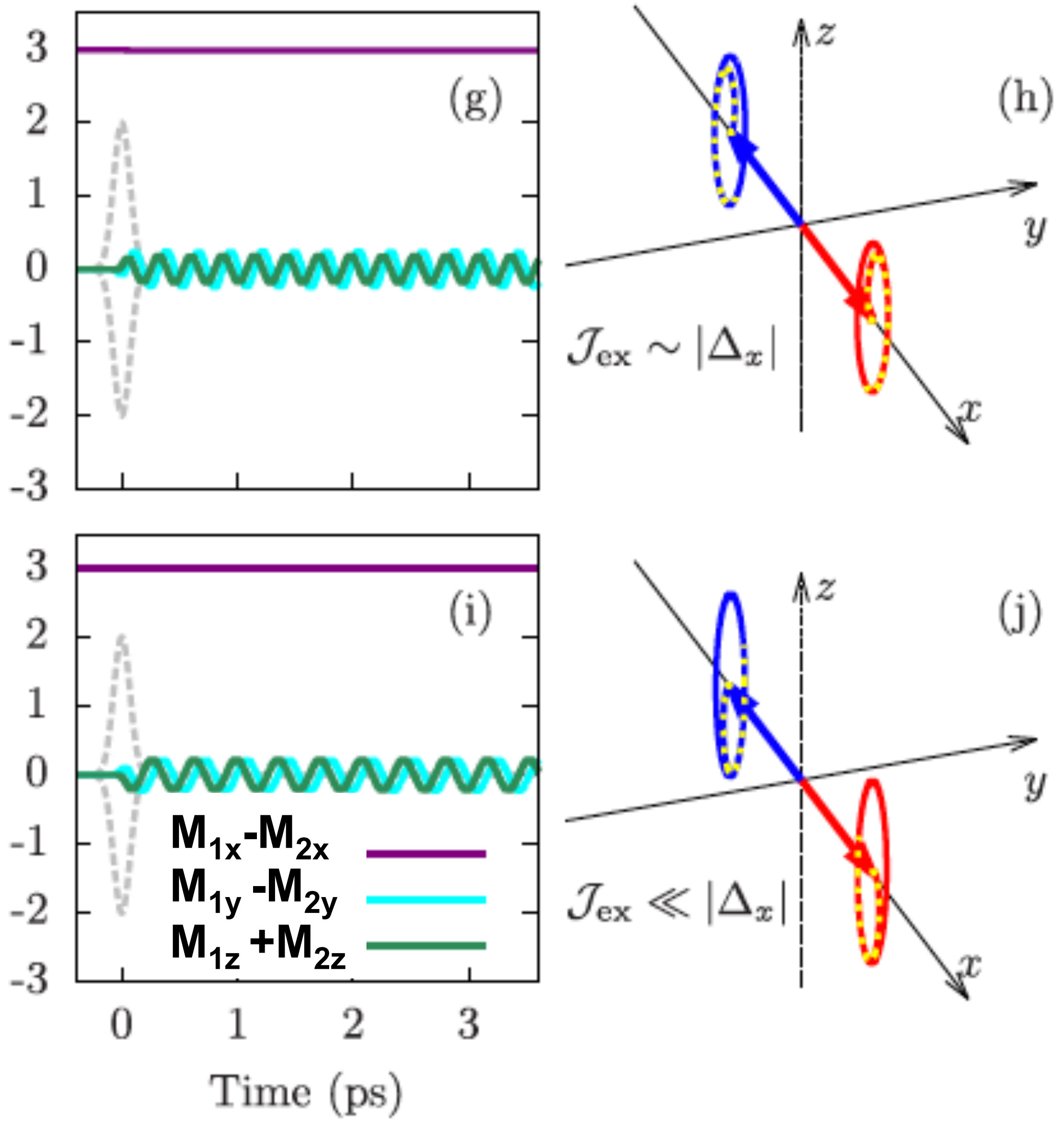
Precession of the antiferromagnet



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Conclusions + outlook

- Even if the pump-pulse duration is 40 times shorter than the period of induced precessions,

  light-induced precessions start during the action of light and affect the position of magnetic vectors after the action of the pump pulse.

- The character of induced magnetic precessions depends on the ratio of the pump-pulse duration to the period of magnetic-oscillation modes

- Explore opportunities to emphasize precessions by adjusting pulse duration or chirping

- Study magnetization dynamics during the action of the pump pulse

Conclusions + outlook

- Study magnetization dynamics during the action of the pump pulse

Few-femtosecond x-ray probe pulse to measure dynamic **during** the driving pulse

Advantage of x rays – atomic selectivity and/or atomic resolution

X-ray scattering provides laser-dressed electron density

Daria Popova-Gorelova, David A. Reis, Robin Santra, Phys Rev B 98, 224302 (2018)

Daria Popova-Gorelova, Robin Santra,arXiv:2012.10334

Daria Popova-Gorelova, V. Guskov, Robin Santra, arXiv:2009.07527

Resonant x-ray probe pulse+ driving pulse – theory under construction

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