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Title: Direct Observation of Magnon Dynamics in Multiferroic HoMnO₃

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Directly probing antiferromagnetic order in HoMnO_3 on an ultrafast time scale

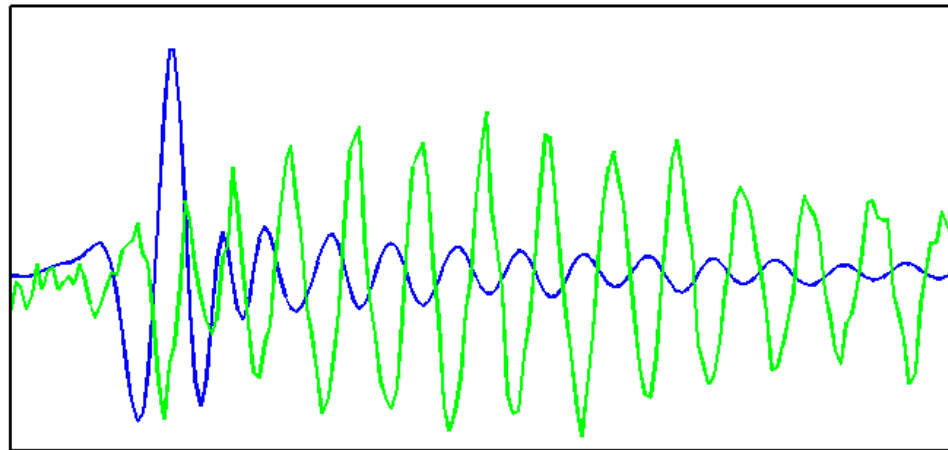
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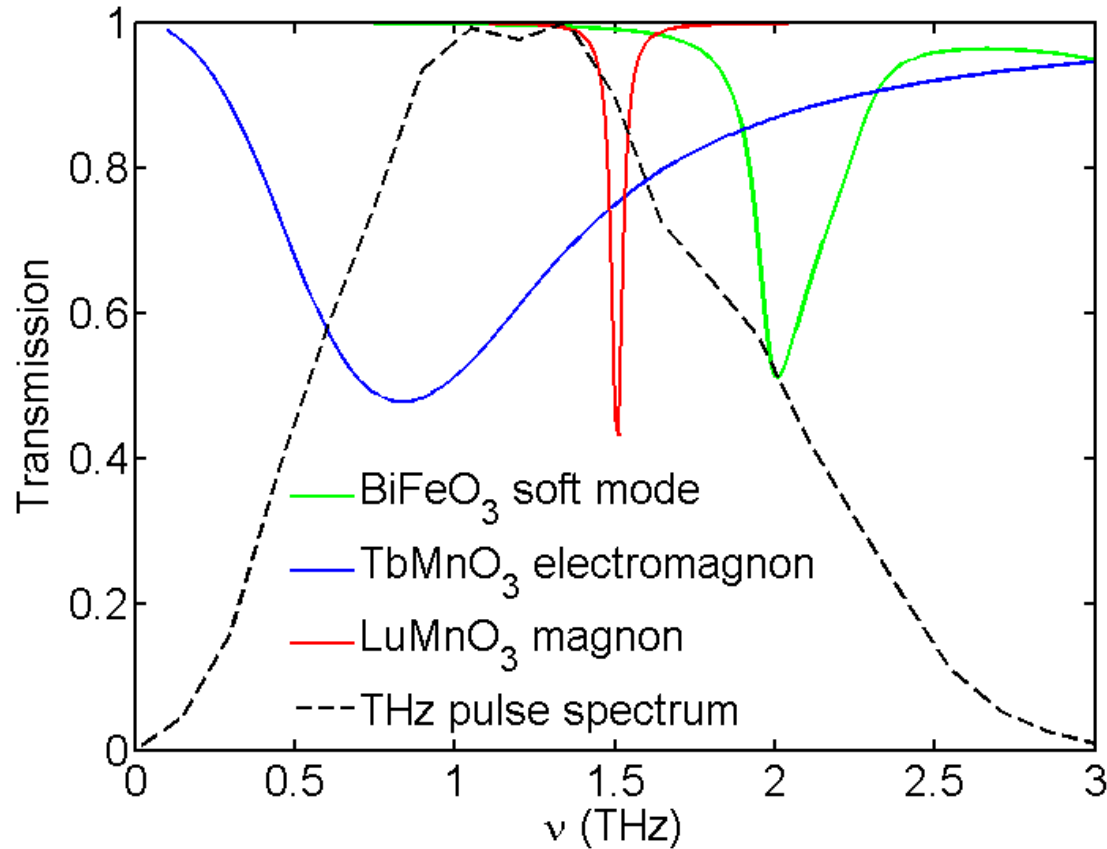
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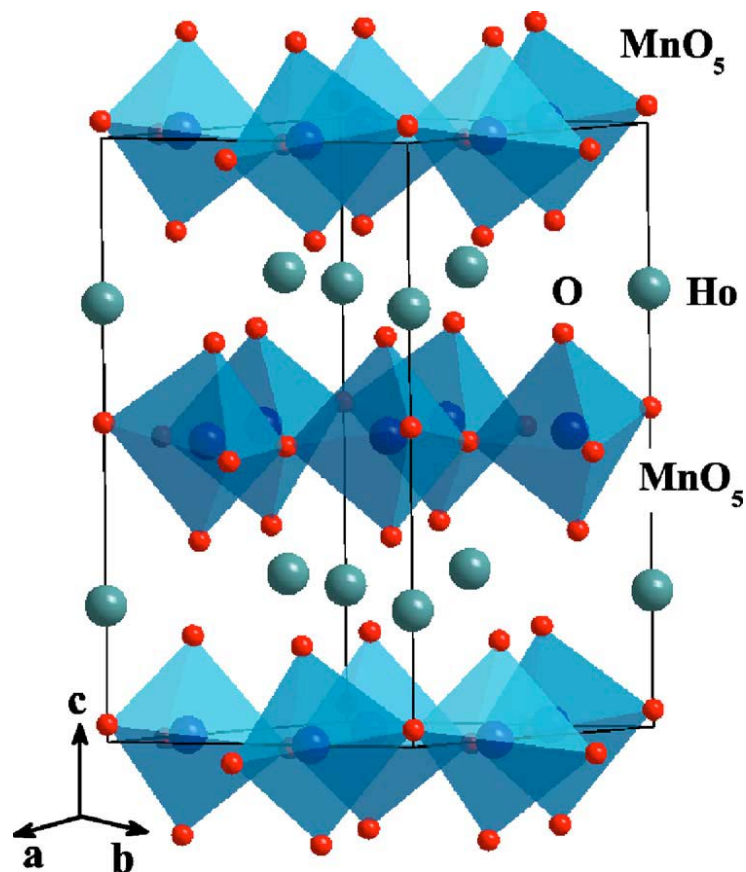
Ultrafast probing of material properties through low energy modes



Ultrashort THz pulses overlap with resonances through which magnetism or ferroelectricity can be probed or controlled.

What is HoMnO_3 ?

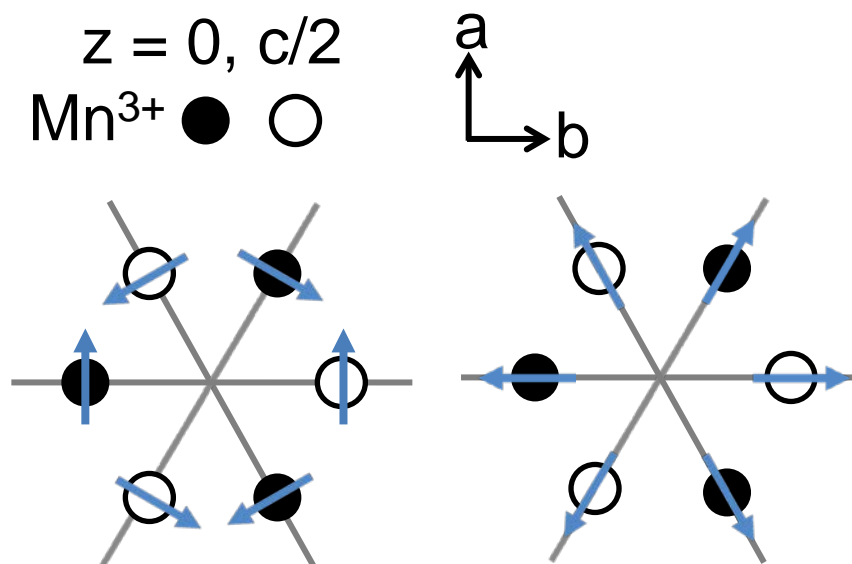
Hexagonal Lattice
 $T \leq T_c \text{ (FE)} = 875 \text{ K}$



Rai, et. al, PRB, **75** (2007).

Frustrated antiferromagnet
 below 78 K

$z = 0, c/2$
 Mn^{3+} ● ○

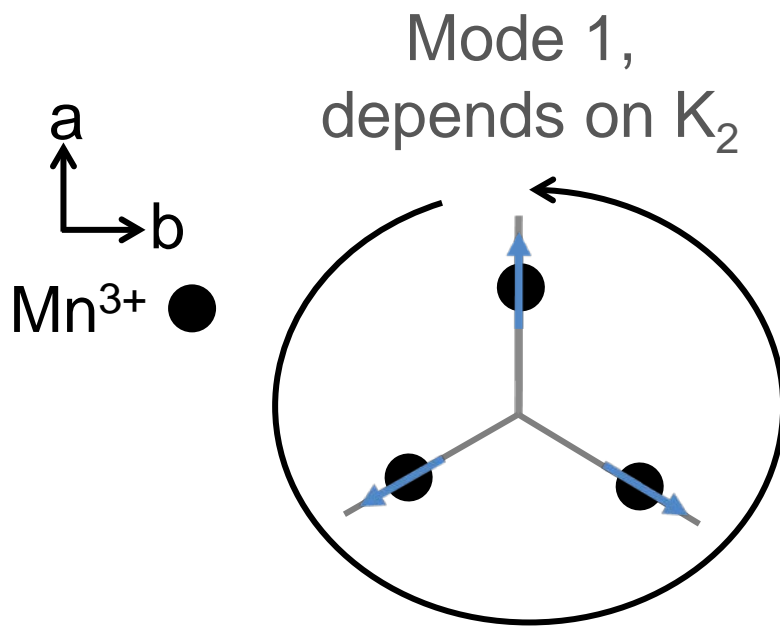


$P\bar{6}_3cm$
 $T < T_{\text{SR}} = 42 \text{ K}$

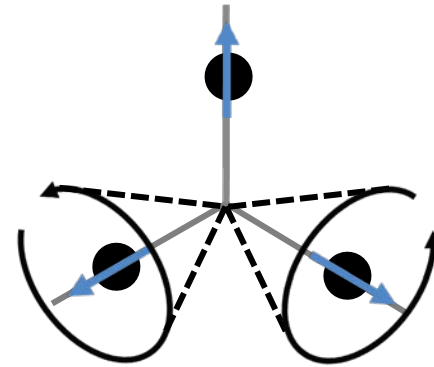
$P\bar{6}_3cm$
 $T < T_{\text{Néel}}$

Fiebig, et. al, JAP, **91** (2002).

Magnons in HoMnO₃



Modes 2 and 3
(degenerate).
Depends on K_1 and λ



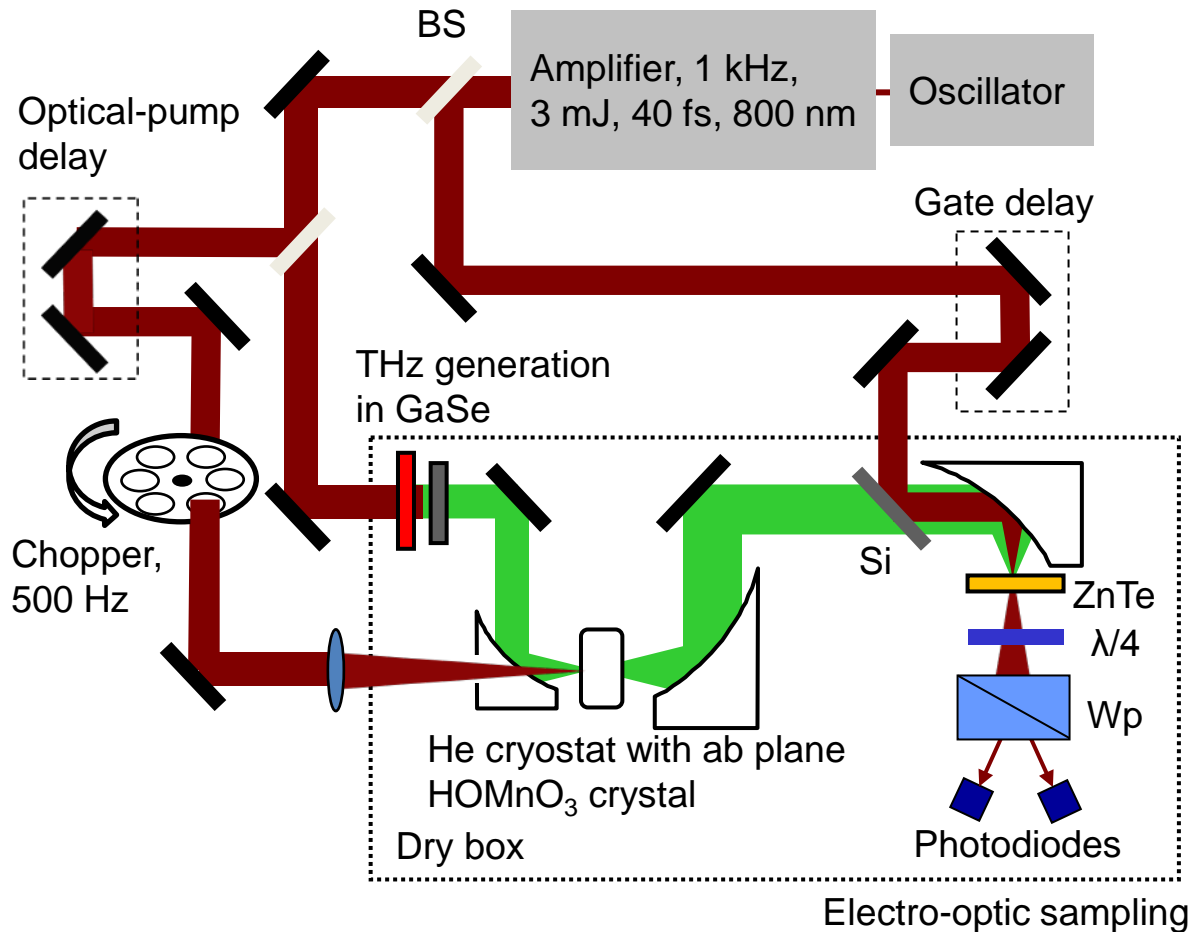
Uniaxial
anisotropy

In-plane
anisotropy

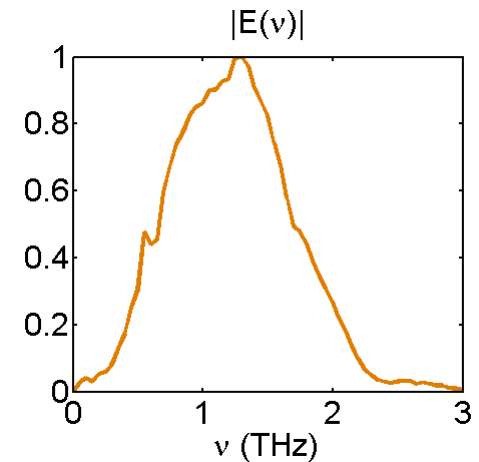
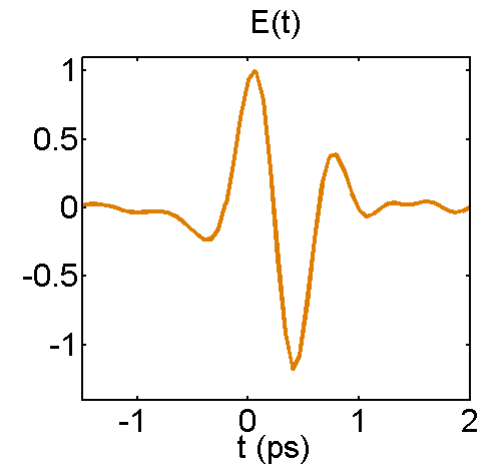
Exchange

$$E = -K_1 \sum_i \cos^2 \theta_i - K_2 \sum_i \cos^2 3\varphi_i + \lambda \sum_{i < j} M_i \cdot M_j$$

Optical-pump, THz-probe experimental setup

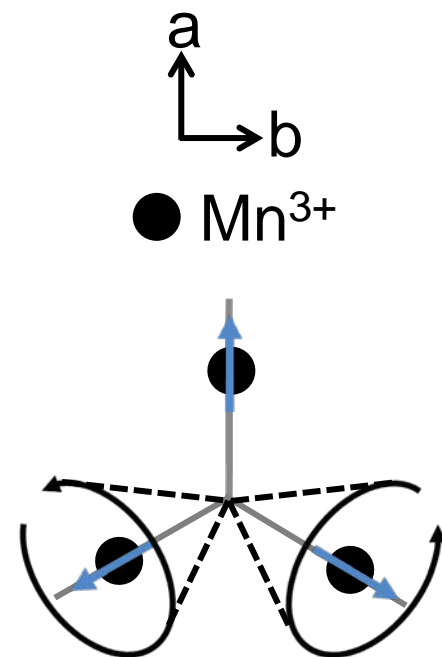
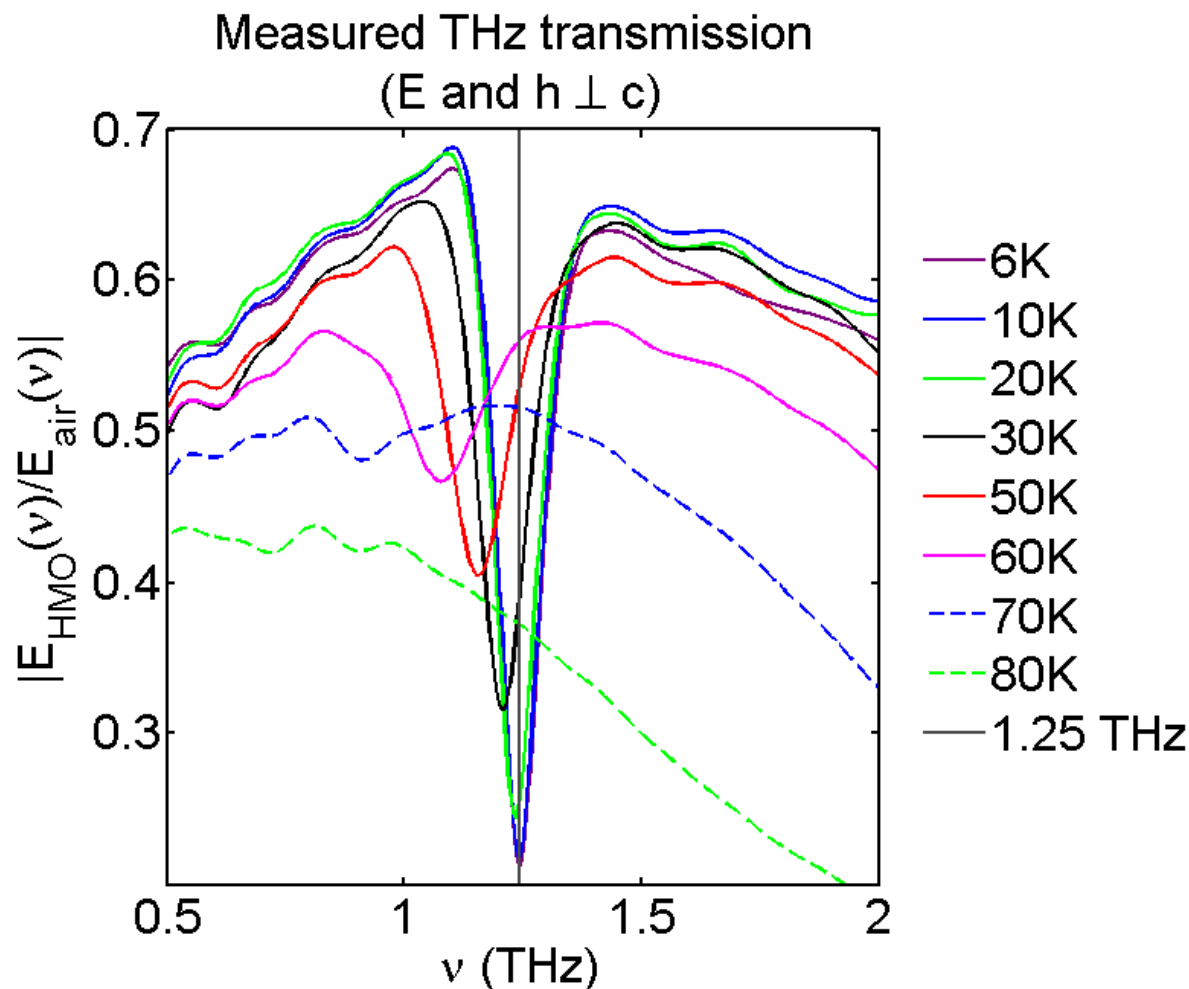


Incident THz pulse



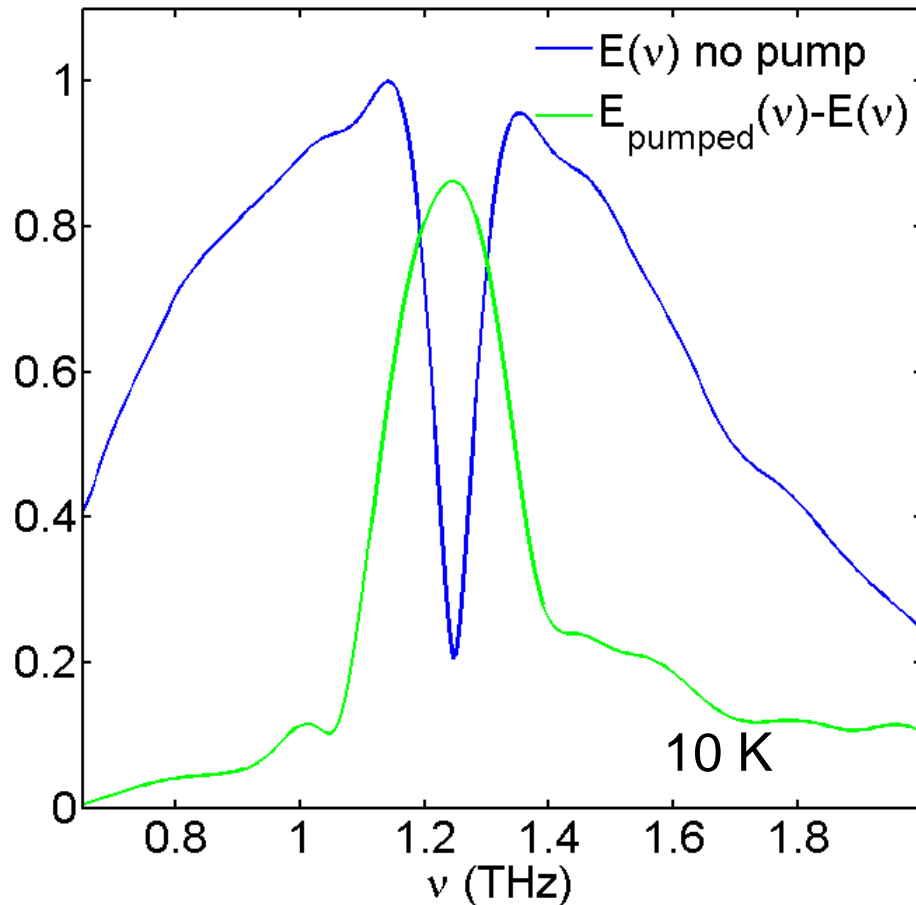
By removing the GaSe crystal and Si filter we can do all optical-pump, probe measurements for comparison.

Steady state THz transmission in HoMnO_3

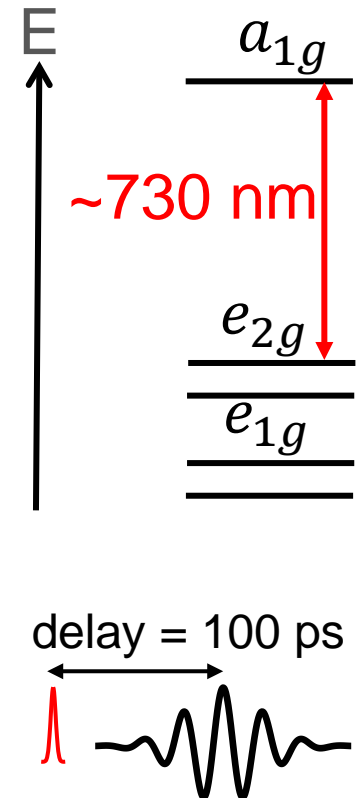


As the temperature increases, the restoring forces and order are weaker, so the mode broadens and lowers in frequency.

Photoexciting HoMnO_3 modifies the magnon



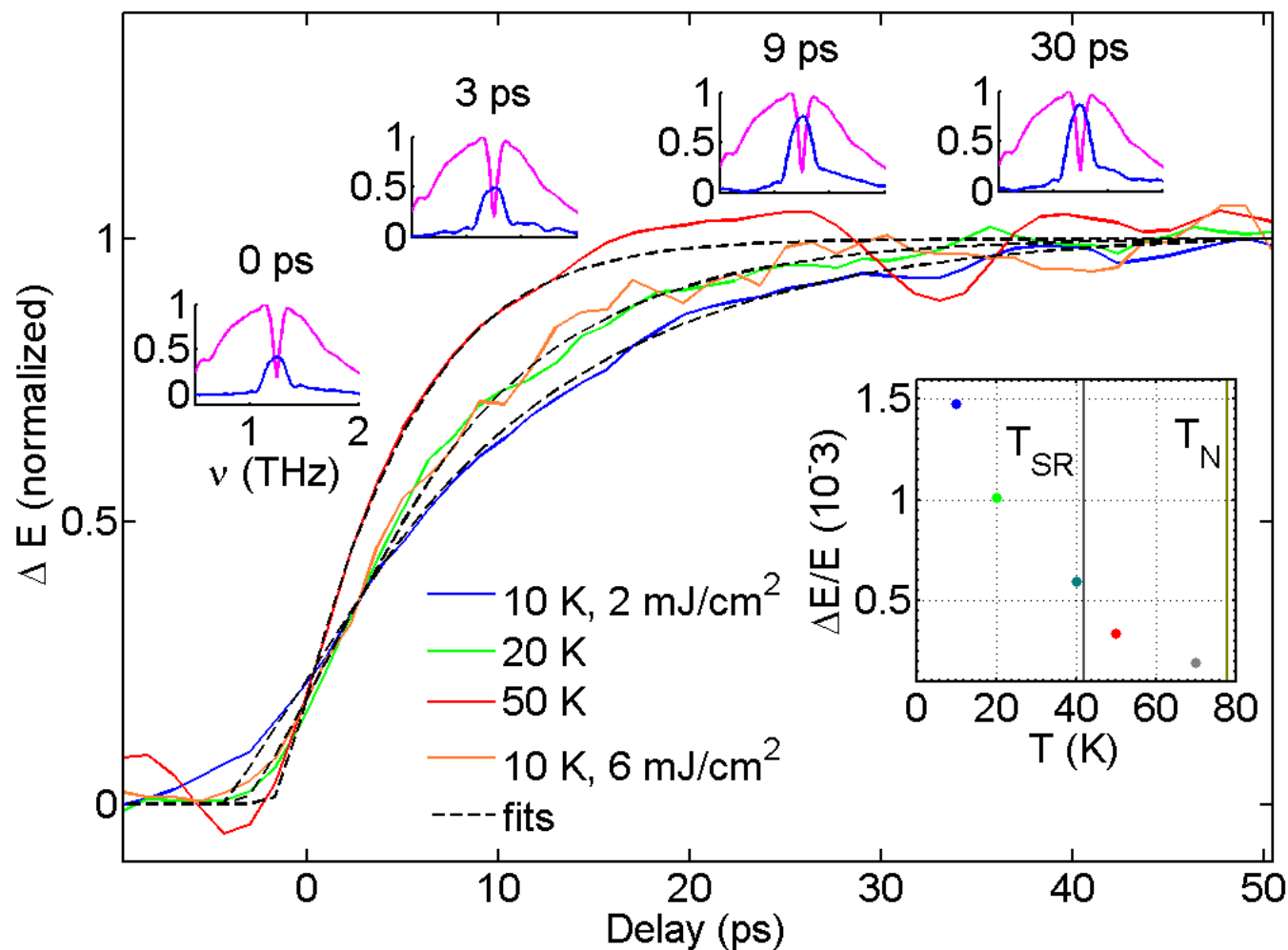
On-site d-d transitions



Optically pumping affects the magnons and not free carriers.

Optical absorption in HMO: Souchkov et. al. PRL **91** 2003 and Rai, et. al, PRB, **75** (2007).

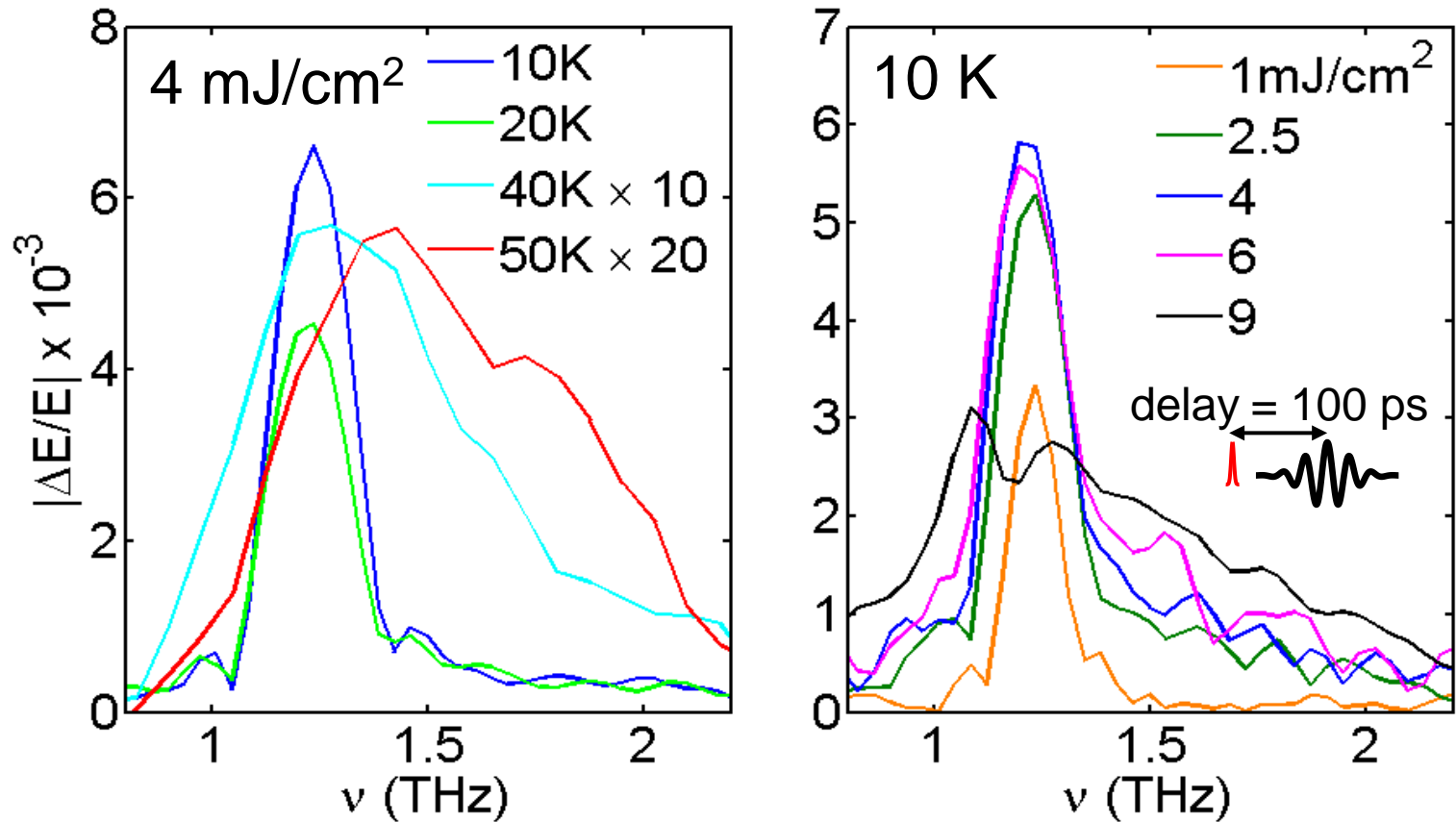
Optically induced transparency vs delay



The signal slowly rises over 5-15 ps. Its amplitude decreases with temperature and is gone by $T_{\text{Néel}}$.

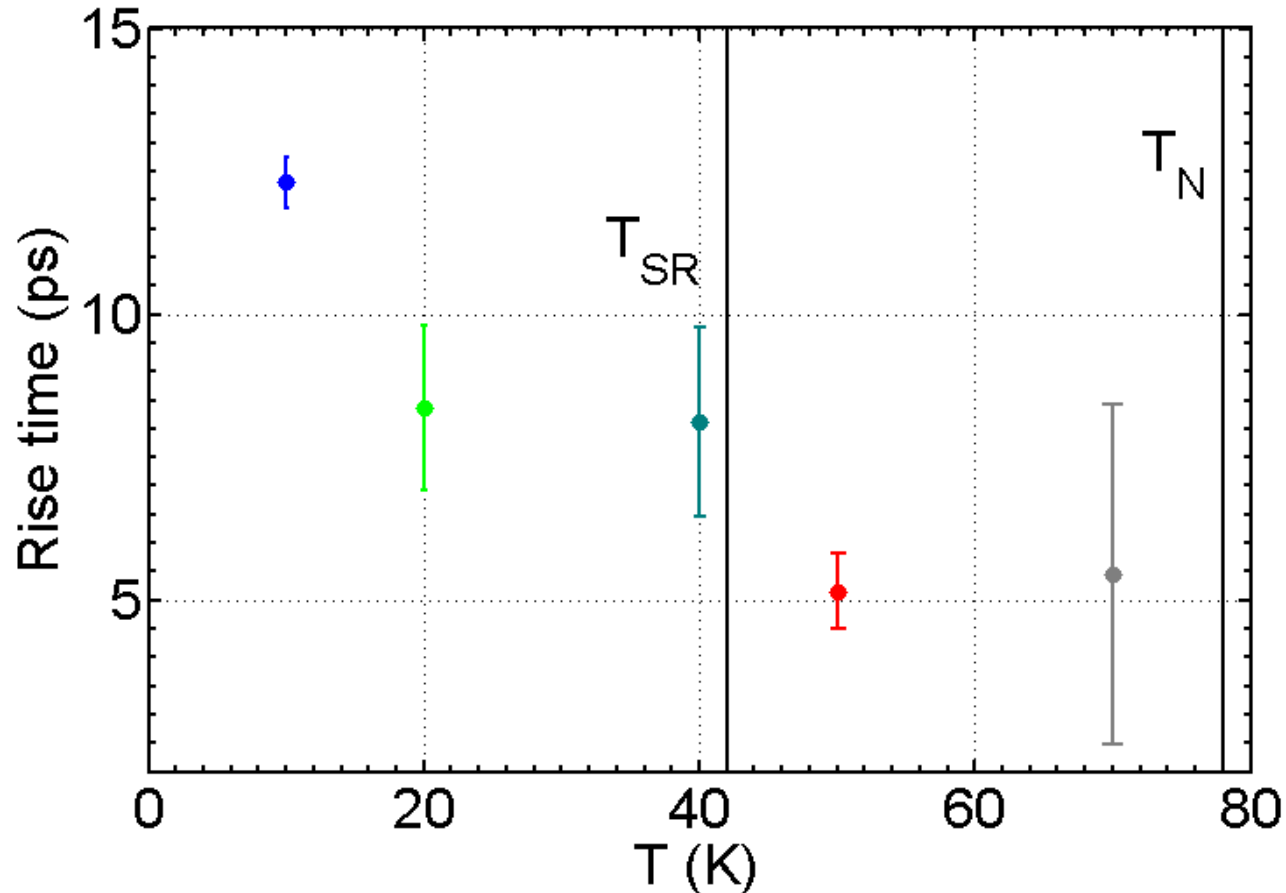
Optically pumping is consistent with steady state heating

Photoinduced changes versus fluence and T



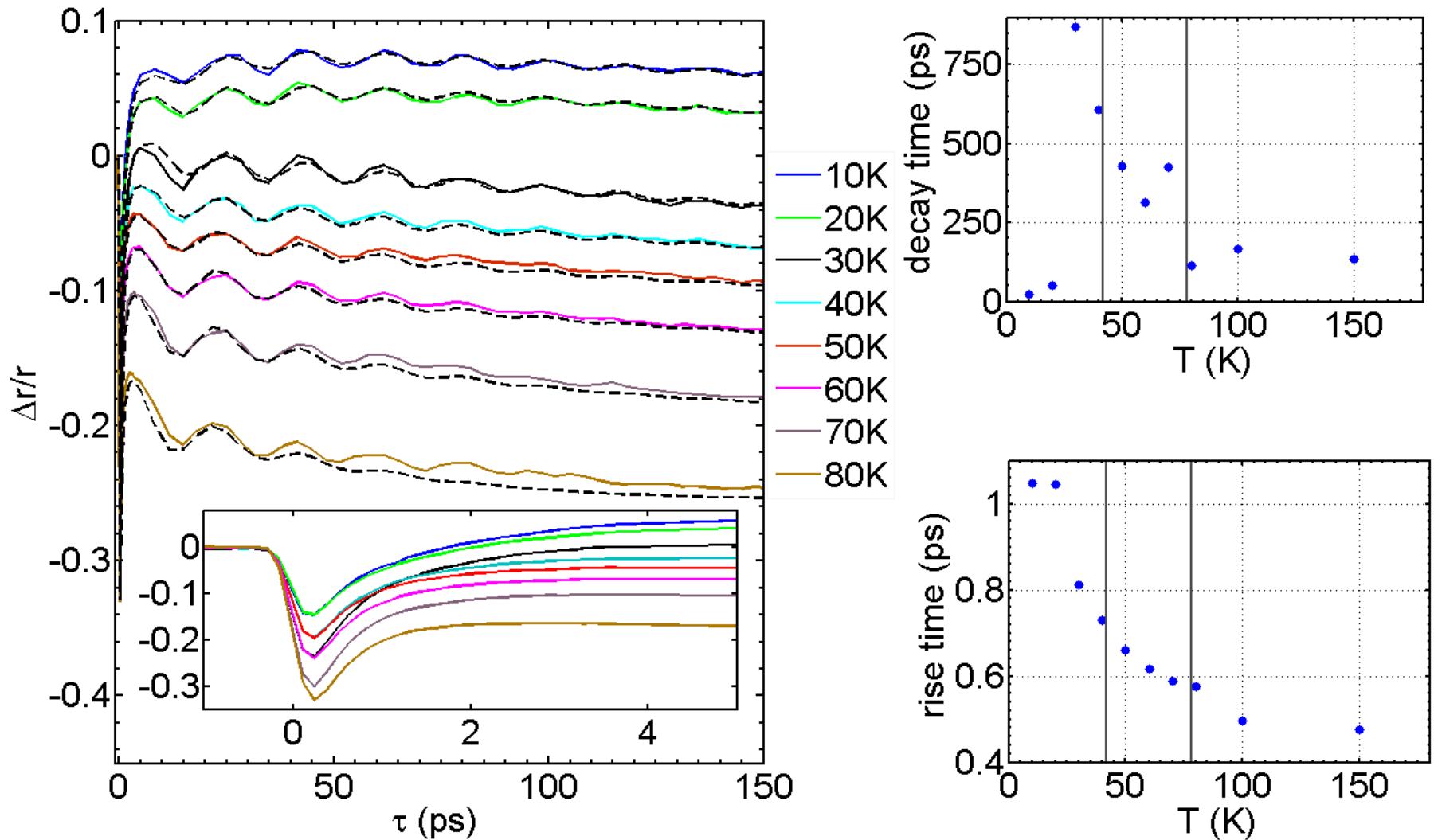
Also, more pumping fluence results in a faster decay time.

Electrons transfer energy to spins via phonons



The faster decay at higher temperature occurs since there are more excited phonons for the spins to gain energy from.

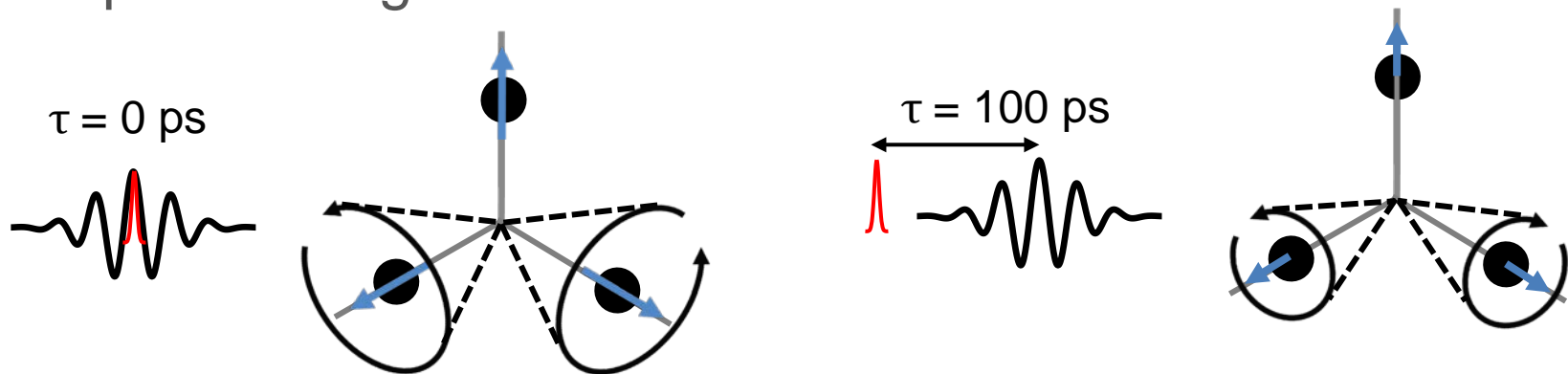
Comparison to probing the optical reflectivity



Unlike the optical reflectivity, THz probes only a single magnon.

Conclusions

- Optically pumping and THz probing HoMnO_3 directly reveals the electron-magnon coupling and its dynamics.
- This occurs on a 5-15 ps time scale and appears like steady state heating indicating that phonons, electrons and spins are all in equilibrium.
- Therefore the electrons transfer their energy to spins via phonons.
- Optical reflectivity probing is very different since it is sensitive to the magnetic order in general and not specific magnon modes as in the THz case.



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