Probing ultrafast photo-induced dynamics of the exchange energy in a Heisenberg antiferromagnet

G. Batignani[1,2], D. Bossini[3], N. Di Palo[1], C. Ferrante[1], E. Pontecorvo[1], G. Cerullo[4], A. Kimel[3], T. Scopigno[1,5]
[1]*Physics Department, University "Sapienza" Rome, Italy,*
[2]*Dipartimento di Scienze Fisiche e Chimiche, Universitá degli Studi dell'Aquila, L'Aquila I-67100, Italy,*
[3]*Radboud University Nijmegen, Institute for Molecules and Materials, Nijmegen, AJ 6525, The Netherlands,*
[4]*Dipartimento di Fisica, Politecnico di Milano, Piazza L. da Vinci 32, Milano 20133, Italy,*
[5]*Center for Life Nano Science @Sapienza, Istituto Italiano di Tecnologia, 295 Viale Regina Elena, Roma I-00161, Italy.*
tullio.scopigno@phys.uniroma1.it

Abstract: An ultrafast enhancement of the exchange interaction between two spins in an antiferromagnetic insulator is detected, developing an all-optical pump–probe method based on stimulated two-magnon excitation.
OCIS codes: (300.6450) Spectroscopy, Raman, (300.6530) Spectroscopy, ultrafast, (290.5910) Scattering, stimulated Raman, (160.3820) Magneto-optical materials

1. Background

Manipulating the macroscopic phases of solids using ultrashort light pulses has resulted in spectacular phenomena, including metal–insulator transitions, superconductivity and subpicosecond modification of magnetic order. The development of this research area strongly depends on the understanding and optical control of fundamental interactions in condensed matter, in particular the exchange interaction [1]. However, disentangling the timescales relevant for the contributions of the exchange interaction and spin dynamics to the exchange energy, E_{ex}, is a challenge. The contribution of the exchange interaction to magnetic dispersion is dominant at the zone edges. A convenient way to access this region is provided by 'two-magnon (2M) light scattering', conventionally described in terms of couples of interacting magnons whose wavevectors lie near the edges of the first Brillouin zone, as determined by the law of conservation of momentum and by the magnon density of states, which typically peaks far from the zone centre.

Spontaneous Raman (SR) spectroscopy has been widely used to measure the frequency of the 2M line, which represents the increase of E_{ex} relative to the ground-state exchange energy, corresponding to a completely ordered spin lattice. Such an increase, in a Heisenberg antiferromagnet, is generated by two spin flip events occurring on neighbouring sites, one for each magnetic sublattice. When SR spectroscopy is used as a probe in time-resolved experiments to address sub-picosecond transient phases it has, however, a critical shortcoming. Specifically, it is subject to a time–energy resolution restriction dictated by the time–bandwidth product of the light pulse, which is limited by $\Delta E \Delta t \geq 15\ cm^{-1}$ ps [2]

2. Results

We present an experimental strategy to circumvent this limitation by taking advantage of the unrestricted time precision and energy resolution of femtosecond stimulated Raman spectroscopy (FSRS), which we use to track the dynamics of the 2M line of the cubic perovskite KNiF3 upon ultrashort optical excitation. This allowed us to tackle one of the basic challenges in femtomagnetism — the direct measurement of the exchange constant between two spins in a magnetic solid on the femtosecond timescale.

When a 60 fs pulse with a photon energy of 1.9 eV and a fluence of 60 mJ cm^{-2} is temporally overlapped as a pump pulse with the probe field, a six-wave mixing process dominates the optical response of the system, albeit with the same magnitude in the Stokes and anti-Stokes lines. Therefore, it can be removed from the time-resolved measurements, undisclosing the transient modification of the two-magnon frequency. After also taking into account possible corrections due to the fact that the dynamics of the two-magnon line is much shorter than the homogeneous lifetime of the Raman transition, the transient modification can finally be attributed to an increase of the exchange energy of 3% completed within 100 fs upon photoexcitation. As KNiF3 is almost transparent at 1.9 eV the observed increase directly reflects the transient modification of the superexchange constant.

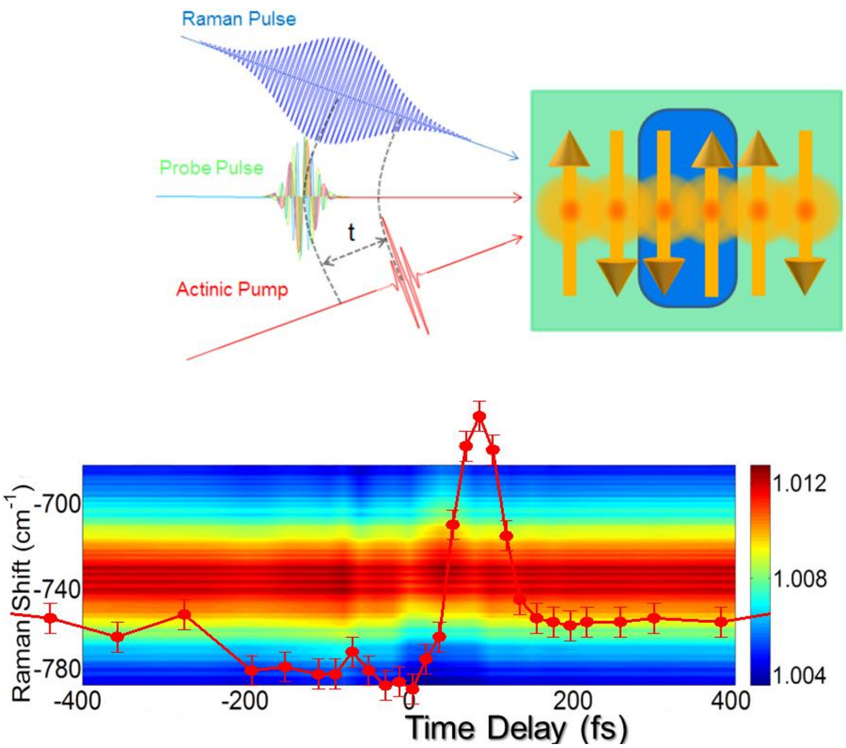

Figure 1: (Top) Sketch ot the experimental beam configuration and of the 2M excitation. (Bottom) Raman profile for different pump-probe delay. Superimposed the 2ML dynamics as determined by the Raman peak position.

3. Methods

We developed a femtosecond broadband stimulated Raman set-up to detect two-magnon scattering, specifically designed to push the time resolution in the relevant femtosecond time domain. In this all-optical set-up, the probe field is given by a femtosecond white-light continuum temporally superposed to a picosecond narrowband Raman pulse. The probe field stimulates the two-magnon transition in $KNiF_3$, which results in gain (Stokes) and loss (anti-Stokes) lines in the white-light continuum transmission spectrum [3], the latter being absent in conventional spontaneous Raman experiments.

A Ti:sapphire laser was used to generate 3.6 mJ, 35 fs pulses at 800 nm, with a 1 kHz repetition rate. A portion of the laser fundamental, driving a two-stage optical parametric amplifier, produced a horizontally polarized 60 fs actinic pump (EA(t)) with 1.9 eV photon energy and 60 mJ cm^{-2} fluence, which was used for photoexcitation. Snapshots of the subsequent 2M line evolution were taken by broadband SRS, where the simultaneous presence of a narrowband picosecond pulse (Raman pulse) and a femtosecond WLC (Raman probe) generates vibrational coherence. Raman pulses were synthesized from a second two-stage optical parametric amplifier producing tunable infrared–visible pulses, followed by a spectral compression stage based on frequency doubling in a 25 mm beta barium borate crystal. Vertically polarized pulses with 2.5 eV photon energy and 10 cm^{-1} bandwidths were obtained, with 5 mJ cm^{-2} fluence. The femtosecond probe comprised a vertically polarized WLC, generated by focusing the laser fundamental into a sapphire crystal. The Raman features arose as gain (positive or negative) on top of the transmitted WLC, which was frequency dispersed by a spectrometer onto a charge-coupled device. A synchronized chopper was used to block alternating Raman pulse pulses in order to obtain the Raman gain using successive probe pulses, and a second chopper blocked the actinic pump at 250 Hz to obtain Raman gain spectra with and without actinic excitation. All the pulses were linearly polarized, the WLC and Raman fields were parallel, and the actinic pump field was rotated by 90° to minimize coherent artefacts.

4. Conclusions

In summary, we have demonstrated that ultrafast coherent Raman spectroscopy can provide experimental evidence for sub-100 fs photo-induced dynamics of magnons in the high-k region of the Brillouin zone, direct expression of a light-induced increase in the exchange energy [4]. Our results provide an estimate of the dissipative dynamics of the 2M line, setting an experimental benchmark for theoretical descriptions of ultrafast laser-induced magnetic dynamics beyond the Heisenberg model. Moreover, we introduce an approach to obtain the real dynamics of a Raman mode from FSRS measurements, on timescales as short as the duration of the pump and probe pulses. We anticipate that an investigation of the dynamics of the 2M mode triggered by resonant excitation of the charge-transfer transition would reveal different pathways for optical control of the exchange energy.

5. References

[1] A. Kirilyuk, A. Kimel and T. Rasing, *Ultrafast optical manipulation of magnetic order*. Rev. Mod. Phys. **82**, 2731–2784 (2010).

[2] C. Rullière, (ed.) Femtosecond Laser Pulses: Principles and Experiments (Springer, 2005)

[3] G. Batignani, E. Pontecorvo, G. Giovannetti, C. Ferrante, G. Fumero and T. Scopigno. *Broadband Stimulated Raman spectroscopy in electronically resonant biomolecules*. Scientific Reports, **6**, 18445, (2016) doi: 10.1038/srep18445

[4] G. Batignani, D. Bossini, N. Di Palo, C. Ferrante, E. Pontecorvo, G. Cerullo, A. Kimel and T. Scopigno. *Probing ultrafast photoinduced dynamics of the exchange energy in a Heisenberg antiferromagnet*. Nature Photonics, **9**, 506, (2015)