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Femtosecond optomagnetism in dielectric antiferromagnets

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Abstract

Optical femtosecond manipulation of magnetic order is attractive for the development of new concepts for ultrafast magnetic recording. Theoretical and experimental investigations in this research area aim at establishing a physical understanding of magnetic media in light-induced non-equilibrium states. Such a quest requires one to adjust the theory of magnetism, since the thermodynamical concepts of elementary excitations and spin alignment determined by the exchange interaction are not applicable on the femtosecond time-scale after the photo-excitation. Here we report some key milestones concerning the femtosecond optical control of spins in dielectric antiferromagnets, whose spin dynamics is by nature faster than that of ferromagnets and can be triggered even without any laser heating. The recent progress of the opto-magnetic effect in the sub-wavelength regime makes this exciting research area even more promising, in terms of both fundamental breakthroughs and technological perspectives.

Keywords: ultrafast spin dynamics, antiferromagnet, dielectric materials, coherent Raman processes

(Some figures may appear in colour only in the online journal)

1. Introduction

In the last two decades a massive research effort has been devoted to the investigation of the interaction between ultra-short laser pulses and magnetic materials [1]. The reason for the increasing popularity of ultrafast spin dynamics lies in the possibility to access and control novel non-equilibrium states of magnets. Since few stimuli other than a laser pulse can perturb a solid on the femtosecond time-scale, an utterly new physical landscape can be investigated in this way. The well-established description of magnetic phenomena based on the concepts of equilibrium and the adiabatic approximation does not apply to the ultrafast regime of spin dynamics.

This research field was opened up by the seminal discovery of the light-induced ultrafast demagnetization [2] of metallic ferromagnets, triggering a discussion about the microscopic mechanism which is not entirely concluded yet. Recent experiments, which investigate the picosecond demagnetization with both time- and space-resolution, have

advanced in a significant way the understanding of this process. A complicated scenario has been unraveled, in which several processes occurring on different time-scales must be considered to fully describe the light-induced picosecond quench of the magnetization [3].

The observation of photo-induced picosecond reversal of the magnetization in ferrimagnetic alloys (all-optical switching) has ignited an even more intense research activity [4]. This process is fully deterministic: a single laser pulse reverses the magnetization and the following pulse switches it back to the initial state. A wide variety of experimental approaches were employed to either achieve a deeper insight in the physics of the reversal, or to create a roadmap towards magnetic-recording applications based on the all-optical switching. In particular, a time- and element-resolved measurement of the spin dynamics of the two antiferromagnetically coupled magnetic sublattices displayed a striking phenomenon: the reversal of the total magnetization is preceded by a transient ferromagnetic state, which is

Figure 1. A Heisenberg antiferromagnet consists of two collinear magnetic sublattices with opposite orientation. Note that the two red arrows ($S^\uparrow$ and $S^\downarrow$) do not represent single spins, but they are the sum of all the spins belonging to the $\uparrow$ and $\downarrow$ sublattices, respectively. The total spin and, therefore, the magnetization are vanishing. It is usual to introduce the antiferromagnetic vector L as defined in the figure. This quantity is the order parameter of an antiferromagnet.

forbidden in a thermodynamical description of a ferrimagnet [5]. Novel theoretical approaches have been developed to describe how the laser-induced non-equilibrium states can produce the exotic phenomena experimentally observed [6, 7]. It is also important to mention that recently the size of the optically switched domain has been shrunk down to 50 nm [8]. This is a remarkable result, since it matches the typical performances of heat-assisted magnetic recording, which will be the next large-scale magnetic recording technology to be released on the market.

As aforementioned, a laser pulse can quench and reverse the magnetization on the ultrafast time-scale. In addition, the idea to control the direction of the magnetization via femto-second laser-induced non-equilibrium states is appealing for technological applications and challenging for the development of fundamental condensed matter physics. The ultrafast modification of the magneto-crystalline anisotropy has been explored by means of a wide variety of approaches: magneto-acoustic coupling [9–11], photo-induced phase transitions [12], ultrafast laser heating of a non-collinear magnetic structure [13], excitation of carriers in magnetic semiconductors [14].

All of the approaches described hitherto have been applied almost exclusively to metals or semiconductors. However, the overwhelming majority of magnetically ordered materials in nature is represented by antiferromagnets. In the most basic approximation, these systems can be described by means of a two-sublattices Heisenberg model, in which the exchange interaction determines an anti-parallel alignment (figure 1).

The study of antiferromagnetic systems is motivated by the naturally higher frequencies of the collective spin excitations, compared to ferromagnets. Consequently, much faster spin dynamics can be triggered and detected, entering the picosecond and even femtosecond regimes. It is crucial to

observe that the substantial majority of antiferromagnets are dielectrics. This fact entails that the visible and near infrared light–matter interaction takes place in a non-dissipative regime, which means that little or virtually no laser-heating occurs. The consequences are profound: coherent Raman-like interaction mechanisms become relevant, given the absence of dissipations. Therefore in this framework we do not consider the spin dynamics arising from non-equilibrium electronic states, which is the typical scenario in metallic systems.

Here we discuss these pathways of light–spin interaction and we show that it allows the excitation, detection and control of coherent collective spin excitations (i.e. spin waves or magnons) in a wide variety of materials. Our discussion is mainly focused on collinear and canted antiferromagnetic structures, although the same approach has been demonstrated in more exotic and complicated spin textures as well (see section 4). Moreover we will report how femtosecond spin dynamics on the nanometer length-scale has been recently accessed. Although this was expected to be possible only by using x-rays, coherent Raman scattering enables visible laser pulses to excite sub-wavelength spin excitations. This result is of great interest also in view of potential applications based on magnonics devices, given the demonstrated manipulations of magnons with nanometer wavelength and femtosecond period.

2. Impulsive stimulated Raman scattering (ISRS)

The interaction between light and a magnetic material is conventionally described in terms of the following Hamiltonian

$$\hat{H} = \sum_{\lambda, \nu} \epsilon^{\lambda\nu} (\hat{S}) E^\lambda E^{*\nu}, \quad (1)$$

where $\epsilon^{\lambda\nu}$ is the dielectric tensor which depends on the spin ($\hat{S}$), while the greek letters are indices and E is the complex amplitude of the electric field of light. The tensor $\epsilon^{\lambda\nu}$ can be expanded in powers of the spin operators as

$$\begin{aligned} \epsilon^{\lambda\nu} = & \sum_i \sum_\gamma K^{\lambda\nu\gamma} \langle \hat{S}_i^\gamma \rangle + \sum_i \sum_\gamma G^{\lambda\nu\gamma\delta} \langle \hat{S}_i^\gamma \rangle \langle \hat{S}_i^\delta \rangle \\ & + \sum_{i,j} \sum_\gamma \rho^{\lambda\nu\gamma\delta} \langle \hat{S}_i^{\gamma\uparrow} \hat{S}_j^{\delta\downarrow} \rangle, \end{aligned} \quad (2)$$

where i, j represent ionic sites and higher order terms are omitted [15]. The tensors in the right-hand side of the equation are in general complex, while all the terms in the Hamiltonian are real [15, 16]. This expansion of the dielectric tensor can be invoked to describe how the properties of a light beam are affected by the interaction with a magnetic material. Therefore the terms in equation (2) represent magneto-optical effects, with different dependences on the spin (linear or quadratic) and with a geometry defined by the components of the magneto-optical tensors. For instance, the first term linear in spin is the Faraday effect, while the second is the Cotton–Mouton effect.

It has been long established that the microscopic mechanism of light–spin interaction responsible for magneto

optical effects is Raman scattering on magnons [15, 17]. Therefore, on a more general level, equation (2) describes the light-scattering on spin excitations, implying that each term in the expansion is related to a specific magnetic mode. More specifically, the first two terms of equation (2) are connected with single spin-flip processes, resulting in the generation of single magnons, which are thus called one-magnon modes. Each spin-flip is a perturbation of the fully aligned state shown in figure 1; therefore the transversal spin components directly describe magnetic excitations, since they vanish in the fully aligned Néel state reported in figure 1. It is important to observe that also the second term of the expansion, despite being quadratic in spin, is linearly dependent on the transversal spin component, in case of high-symmetry materials [15]. Although both terms are associated with the generation of one-magnon modes, the selection rules are in general different, since different tensors are involved. The last term in the expansion of $\epsilon^{\lambda\nu}$ is significantly different, since it is quadratic in the transversal spin components, thus describing the generation of two magnons. This term will be analyzed in section 6.

The possibility to access Raman-active lattice modes employing laser pulses via the ISRS mechanism has been widely exploited in condensed matter physics [18, 19]. Two frequency components of a laser pulse can induce a phonon via a stimulated Raman scattering process. This implies that the difference in frequency of the two relevant components must match the phonon frequency. Since ultrashort laser pulses are intrinsically broadband, several pairs of frequency components can give rise to such a process. Translating this condition in the time-domain, it follows that the pulse duration must be shorter than the period of the Raman-active mode. A further requirement to fulfill for a successful ISRS excitations relates to the polarization of the laser beam, which must follow the Raman selection rules. An analogous process involving magnons has been recently demonstrated [20] and it was properly defined as an *opto-magnetic effect*. In fact, the generation of magnons via ISRS is in essence a light-induced modification of the magnetic state of a medium. This is the reciprocal effect of the magneto-optical effects, which justifies the name opto-magnetism. Note that the ISRS process is significantly different in comparison with spontaneous Raman scattering: the light induced magnons are fully coherent, because of the constant relative phase between the frequency components active in the ISRS process. Consequently, in a time-resolved experiment the macroscopic phase of the coherent magnon can be directly visualized and manipulated.

In the following sections we will revise the most significant results of the ISRS excitation of one-magnon modes.

3. Fundamentals of ISRS excitation of one-magnon modes

The Raman excitation of magnons is a very general concept, not restricted to a specific class of materials or magnetic structures. In this section we will introduce the most relevant

Figure 2. The dispersion of a Heisenberg antiferromagnet was calculated by following a standard procedure [22]. In this particular case we considered the cubic system KNiF_3 , whose anisotropy and exchange fields are well known in the literature [23]. The region in which the short-range interactions dominate is marked by the blue area: here the spin excitations have femtosecond period and nanometer wavelength.

results, which explore different important aspects of the spin dynamics triggered by one-magnon modes.

Let us start by recalling some fundamental conservation laws underlying the Raman process. The conservation of energy is taken into account, since the pairs of frequency components in the pump pulse involved in the ISRS are separated by the frequency of the photo-generated magnon. On the other hand, the conservation of momentum limits the optical stimulus to access only spin excitations with almost vanishing wavevector. The typical dispersion of magnons in an antiferromagnet (see figure 2) shows that the lowest-energy spin waves have wavevectors at the center of the Brillouin zone. In this region of the dispersion, the spin-orbit coupling is the most relevant magnetic interaction [15]. Although the theory of light-scattering on magnons demonstrates this fact rigorously [15, 21], a simple observation on the characteristic length-scale gives the justification of this statement. The wavelength of low-energy magnons is on the micrometer scale, which matches the typical scale of long-range interactions like the spin-orbit coupling and Dzyaloshinskii–Moriya interaction [15, 21]. Short-range interactions, like the Heisenberg exchange, become relevant for high-frequency and shorter-wavelength magnons (see section 6).

The original breakthrough observation of the ISRS excitation of magnons was achieved in the canted antiferromagnet DyFeO_3 [20]. Circularly polarized laser pulses were able to trigger coherent magnons, the subsequent macroscopic oscillations of the magnetization were detected by measuring the transient Faraday rotation. It is remarkable that both the excitation process and the detection scheme relies on the first term appearing in equation (2). For this reason, this specific excitation mechanism was named inverse Faraday effect. Note that the coherent nature of the photo-generated spin waves allowed to control the macroscopic phase of the ensemble by changing the helicity of the laser pulses (see figure 3). The photon-energy of the pump beam (1.5 eV) lies in the bandgap of DyFeO_3 , therefore only minor dissipation

Figure 3. Magnetic excitations in DyFeO₃ probed by the magneto-optical Faraday effect. The circularly polarized pump beams of opposite helicities excite oscillations of opposite phase. Inset shows the geometry of the experiment. Vectors δH^+ and δH^- represent the effective magnetic fields induced by right-handed σ^+ and left-handed σ^- circularly polarized pumps, respectively. Reprinted by permission from Macmillan Publishers Ltd: Nature [20]. Copyright 2005.

due to localized d–d transitions occurs and the light–matter interaction regime is definitely non-dissipative.

An analogous excitation, relying on the second term in equation (2), was reported in the canted antiferromagnet FeBO₃ [24, 25]. The requirements in terms of polarization of the pump beam are dictated by the tensor components of the Cotton–Mouton effect. The observed excitation was therefore defined inverse Cotton–Mouton effect.

The experiments described hitherto were performed on materials with canted sublattices, which generate a non-vanishing magnetization in the ground state. This fact raised a fundamental question about the role of the magnetization in the ISRS process: is it at all necessary? Moreover, some doubts concerning the generality of this approach naturally arose, since only materials with huge spin–orbit couplings were previously investigated. The problem was tackled by performing a magneto-optical pump–probe experiment in the fully compensated antiferromagnet NiO [26]. The magnetic structure of this medium can be described by the Néel state shown in figure 1. The photo-excitation triggered two different coherent magnon modes, demonstrating experimentally that the presence of the magnetization in the ground state is not required for a successful ISRS generation of magnons. A later investigation of the same compound revealed even the optical excitation of a third Raman-active magnon mode [27]. The crystal field in NiO quenches the orbital angular momentum, which thus entails that the spin–orbit coupling in the ground state is absent. The successful experiments in NiO confirmed that only the spin–orbit coupling in the excited state is relevant. Therefore ISRS allows to trigger coherent magnons even in magnets with no spin–orbit coupling in the ground state, provided that it does not identically vanish in the excited state. This condition is indeed not tight, given the

strong perturbation of the rich electronic structures of dielectric materials induced by an optical femtosecond laser pulse.

Although experiments had established that the ISRS mechanism applies to several classes of magnetic materials, this approach appeared to be intrinsically hampered since light-induced heating was always present and the cooling dynamics could limit the speed of spin manipulation. As aforementioned, although dielectric materials were optically pumped with photon energies tuned into the energy gap, some local energy dissipation indeed occurred due to the excitation of d-electrons. This criticism called for an experimental investigation, in which spin dynamics could be alternatively excited in the presence and absence of laser heating. Therefore it is also crucial to select the proper magneto-optical effect to probe the spin dynamics. If a spin precession is triggered in an antiferromagnet a non-zero Faraday rotation is indeed measured, because a transient net magnetic moment proportional to $\gamma \vec{L} \times \partial \vec{L} / \partial t$ appears in the non-equilibrium state [28]. However, the Faraday effect cannot detect heating-induced dynamics, since it produces incoherent magnons with no-phase relation with each other. A change of the longitudinal sublattice magnetization in an antiferromagnet does not give rise to a modification of the magnetization (the cross product is zero) but it changes the antiferromagnetic vector. Therefore, a magneto-optical effect other than the Faraday rotation must be probed, in order to discuss the laser-induced heating processes.

The dielectric antiferromagnet KNiF₃ is an excellent candidate to address the aforementioned issues. It is characterized by a region of nearly zero absorption in the optical range, as shown in figure 4(a). A wide energy band gap ($E_g \approx 6.2$ eV [30]) distinguishes this material from 3d transition-metal magnetic oxides, whose energy gap is typically $E_g \approx 3$ –4 eV [31, 32]. All the absorption bands visible in the spectrum are due to relatively weak phonon-assisted localized d–d transitions [30, 33]. Of interest for our purpose, in the 1.5–3 eV range these features alternate with a transparency window around 2 eV, where the absorption coefficient is nearly-zero. KNiF₃ has a perovskite type crystal structure (point group m3m) [34]. Two equivalent Ni²⁺ sublattices are antiferromagnetically coupled below the Néel temperature $T_N = 246$ K [35]. This material is known as a Heisenberg antiferromagnet because of its very weak cubic magnetic anisotropy. The positive sign of the anisotropy constant determines the alignment of spins along the [001], [010] and [100] axes [16]. The magnetic structure is described in terms of the antiferromagnetic vector, defined in figure 1. The quantity experimentally detected was the modification of the ellipticity of the probe beam, which in our experimental configuration (see figure 4(b)) represents variations of the magnetic linear birefringence (Cotton–Mouton effect). In a Heisenberg antiferromagnet this effect is proportional to all the possible quadratic combinations of the spin and thus of $\vec{L}$ [36]. Measuring the transient magnetic linear birefringence allows to detect simultaneously both the transversal and the longitudinal dynamics of $\vec{L}$ [29, 36].

Figure 4. (a) The absorption spectrum of KNiF₃ was measured at room temperature with a xenon lamp. The photon energies shown in the figure were employed in [29]. (b) This experimental geometry allows the measurement of transient magneto-optical effects in a pump–probe scheme. The dynamical rotation [22] or the ellipticity [29] of the probe polarization as a function of the pump–probe delay Δt were measured.

A spectroscopic investigation of the opto-magnetic effect was performed tuning the photon energy in the range of 1.5–3 eV, either to one of the absorption bands or to the transparency window, as shown in figure 4(a). Two different scenarios can thus be accessed (see figure 5). If an electronic transition is excited, a fraction of photons in the pump pulse participates in the generation of the coherent magnons via ISRS, while another fraction is absorbed, leading to a population of the upper localized 3d band (see figure 5(a)). These excited electrons relax on a time-scale of about 1 ps via non-radiative processes, governed by electron–phonon coupling. As the temperature of the phonons T_{ph} increases, the excess energy is exchanged with the spin system on the ≈ 100 ps time-scale due to magnon–phonon coupling, resulting in an increase of the magnon temperature T_M . In this case, the data revealed two different time scales for the relaxation processes of the magnetic system: the damping of the coherent oscillations (magnon scattering) and the increase of the incoherent background (magnon–phonon scattering) [29]. The harmonic component of the signal represent the transversal spin

Figure 5. (a) In the dissipative light–matter interaction regime, the photo-excitation triggers an energy transfer involving the electrons, the lattice and the spins, eventually resulting in heating of the spin system. The coherent generation of magnons via ISRS occurs as well. (b) If the pump photon energy is tuned to the transparency region, only the ISRS process is photoinduced. The damping of the coherent magnons, which is due to scattering events, is the only source of heating for the spin system.

dynamics, while it was demonstrated that the background results from the longitudinal components [29]. Since the two interactions responsible for these two decay processes are different, the time-scales are of course different.

Let us now consider the laser-induced processes occurring when the medium is transparent for the pump pulse, as shown in figure 5(b). The data in [29] show that, unlike in the case of any other photon energy, the damping of the oscillations takes place on the very same time-scale as the dynamics of the incoherent background. Since no real electronic transitions are excited, the energy deposited by the pump laser pulses into the medium is entirely stored in the magnetic ensemble, via ISRS. The laser-generated coherent magnons scatter with a characteristic time ≈ 40 ps, bringing the ensemble of these quasiparticles to decoherence. As a result, the spin temperature increases, traced by the longitudinal dynamics of the antiferromagnetic vector, with the very same characteristic time ≈ 40 ps. This equality proves that, by tuning the energy of the pump photons to the nearly-zero-absorption spectral range, we achieved a regime in which the light-induced spin dynamics does not reveal any signatures of heated electrons or phonons, as if a selective excitation of spins only occurred. This novel regime of spin dynamics has been named *zero-absorption regime* [29].

The experiments discussed in this section provide all the fundamental aspects of the ISRS excitation of one-magnon modes. In fact they demonstrate that this concept is very general and applicable to several magnetic structures and classes of materials (rare-earth orthoferrites, borates, trifluorides and transition metal oxides). Moreover, the requirement of spin–orbit coupling only in the excited state has been demonstrated, as well as the possibility to trigger coherent magnons without any electronic and lattice heating. The nature of this excitation mechanism is purely non-dissipative.

From the point of view of theory, it is worth to remind that the general theory of light-scattering on magnons has been adapted to the impulsive case [37] to describe the excitation mechanism. The modeling of the spin dynamics subsequent the photo-generation of magnons has been formulated relying on classical Landau–Lifshitz-like equations of motion [37–39]. This approach is successful because a single spin-flip process induces a macroscopic precession of spins, which can be described in a phenomenological way as induced by an effective field representing the action of light [39, 40].

After having presented the fundamentals of the ISRS process on one-magnon modes via a series of experiments, we proceed in the next session by reporting novel approaches and trends which further explore the spin dynamics induced by the one-magnon mode.

4. Recent developments of ISRS excitation of the one-magnon mode

The idea of triggering optically coherent magnons in the THz regime in the absence of laser-heating is appealing for the perspectives of magnon spintronics. This research field aims at utilizing the spin currents carried by magnons to perform logical operations [41]. The all-optical approach has the further advantage to be contact-free, i.e. no electric contacts are required on the sample.

The challenges to face for the development of a magnon-based technology are the generation, detection and control of the spin waves. The experiments discussed in the previous section demonstrated the all-optical generation and detection of coherent spin waves. Although the polarization of the pump beam could modify the macroscopic phase of the magnons, an arbitrary control of the amplitude of the magnons was not demonstrated.

A coherent manipulation of light-induced spin waves can be achieved by exciting the magnetic material with a series of delayed identical pump pulses. If the delay is equal to the period (or a multiple) of the magnon the oscillations are enhanced; while they are quenched if the delay among the pump pulses is set to a half period (or an odd multiple) of the magnons. Practically, this concept was demonstrated employing two pump pulses for the control of the photo-excited magnons in DyFeO_3 [40] (see figure 6).

A step further was taken in an investigation of the pure antiferromagnet NiO . First, the coherent manipulation of the magnon amplitude was shown, using the same excitation scheme reported here above but with a different detection. The spin dynamics was probed by monitoring the emission of THz radiation from the light-induced spin waves. A difference in the polarization direction was then introduced between the two linearly polarized pump pulses. Tuning this polarization difference and the delay between the pulses, the motion of the magnetization vector was manipulated to follow an arbitrarily designed direction and amplitude. The trajectory of the oscillating magnetization in the three-dimensional

Figure 6. (a) Precession triggered by the first laser pulse. (b) Amplification of spin precession by the second laser pulse that comes with a delay equal to a multiple of full periods. (c) Quenching of the spin oscillations by the second pump that comes with a delay equal to an odd multiple of half periods. Figure from [40]. Copyright Wiley-VCH Verlag GmbH & Co. KGaA. Reproduced with permission.

space was then reconstructed, via the THz emission measurement of all the components [42].

Another relevant result towards magnonics application was the establishment of the *optomagnonics* concept. In a magneto-optical time-resolved experiment, the photo-excitation of the antiferromagnet YMnO_3 revealed a complicated dynamics of the ellipticity of the polarization. Several magnetic and lattice modes were optically excited and detected. On the one hand, changing the polarization of the pump beam allowed to selectively excite single-magnon modes. On the other hand, even if several modes were triggered, a detection sensitive to a single mode was established [43]. This concept allows for the transfer of the information contained in the polarization eigenstate of a fully polarized pump beam into the magnetic eigenmodes in a one-to-one fashion. The information was then converted back from the magnetic system to the polarization state of a probe beam, once again in a one-to-one process (see figure 7). This write-read cycle is the proof of concept of the feasibility to employ laser pulses as a stimulus in magnonics, i.e. the *optomagnonics*.

All the experiments reported so far investigated the local spin dynamics, since the pump and probe beam were always overlapping. This scheme does not allow to study the

Figure 7. Illustration of one-to-one information transfer light $\rightarrow$ magnetism $\rightarrow$ light in hexagonal YMnO_3 . The one-to-one mapping between the Stokes parameters of the pump pulse and the magnetic oscillation modes of YMnO_3 , and between these oscillation modes and the Stokes parameters of the probe pulse (prime symbol), is shown. A probe pulse component S'_i is modified by a magnetic oscillation along the j -axis (resulting from a pump pulse component S_j) by an oscillation $\Delta S'_i$ with ijk according to the Levi-Civita tensor ϵ_{ijk} . Reprinted by permission from Macmillan Publishers Ltd: Nature Photonics [43]. Copyright 2005.

propagation of the light-induced spin waves. Several imaging schemes which combine femtosecond time-resolution and micrometer space-resolution have been suggested [45, 46]. Among these, we would like to report the results obtained by scanning the probe beam across a specimen of a ferrimagnetic insulator [44]. An ISRS excitation of magnons was observed, followed by the time- and space-resolved dynamics of the spin system. In particular, the dependence of the signal on the spot size of the focussed pump beam was studied. If the spot size is as large as 2.5 mm, then mostly spin waves with wavevectors close to the center of the Brillouin zone were excited. This magnetic mode corresponds to the ferromagnetic resonance, which causes homogeneous precession of the magnetization. Focussing the pump beam to a smaller spot (down to 25 μm), it is possible to excite higher-wavevector spin waves. Moreover a control on the directional emission and propagation of spin waves was achieved via a pulse shaping of the pump beam (see figure 8). Analyzing the pump distribution in the framework of Fourier theory, it was possible to reproduce time- and space-resolved magneto-optical images with an extremely high degree of accuracy.

We would like to conclude this section, by mentioning some recent works about ISRS generation of magnons in more complicated magnetic structures. In the previous section we pointed out that the one-magnon mode is generated by a light-induced modification of the spin-orbit coupling. However, in magnetic materials which cannot be properly described by the Heisenberg model, other interactions can give rise to the generation of one-magnon modes. For instance, it has been recently demonstrated that an impulsive modification of d electron concentration unbalances the d-f exchange interaction and results in the generation of one-magnon modes in rare-earth chalcogenides [47]. The spin wave associated with the modulation of the magnetic moment induce coherent oscillations of the bandgap energy, detectable

via a measurement of the transient-reflectivity. In a wide variety of canted antiferromagnets, a light-induced perturbation of the ratio between the exchange interaction and the Dzyaloshinskii-Moriya interaction may result in the generation of one-magnon modes [48]. The excitation mechanisms reported in these two works are analogous to the ISRS, with the difference that interactions other than the spin-orbit coupling are relevant in the photo-induced excited state.

In the last few years the research community investigating magnetic phenomena has devoted a very intense interest in topological structures, like skyrmions. It was shown that femtosecond laser pulses can induce similar structures, i.e. bubble domains [49], and even drive them [46]. Here we would like to report a pioneering investigation about dynamics of skyrmions induced via ISRS. The dielectric material Cu_2OSeO_3 exhibits a rich phase-diagram, with a ferrimagnetic, a conical and a skyrmion lattice phase. Exciting the material in the transparency region in the skyrmion lattice ground state, dynamics of the skyrmions were photo-induced. In particular, changing the polarization of light and the experimental geometry (i.e. the direction of the magnetic field) all the skyrmion modes were excited (rotational clockwise, rotational counterclockwise and breathing) [50]. Although the investigation of photo-induced dynamics of skyrmions and other topological structures is only in the initial stage, the potential to manipulate them on the ultrafast time-scale and without heat dissipation is unique to the optical stimulus.

In the next section we will describe how the ISRS mechanism can allow to excite the intrinsically highest-energy and shortest-wavelength magnons. This results in pushing the opto-magnetic concept to the extreme in terms of characteristic time- and length-scales.

Figure 8. Two-dimensional maps of spin wave emitted by a spatially shaped light pulse. (a) Pumping geometry with a rectangular aperture (6 mm $\times$ 1.2 mm). Two-dimensional maps of spin-wave emission obtained by experiment (b) and calculation (c). The longer side of the aperture is parallel to the x -axis. (d) Amplitude map of spin-wave emission extracted from (c). Two-dimensional maps of spin-wave emission obtained by experiment (e) and calculation (f). The longer side of the aperture is parallel to the y -axis. (g) Amplitude map of spin-wave emission extracted from (f). Reprinted by permission from Macmillan Publishers Ltd: Nature Photonics [44]. Copyright 2012.

5. ISRS excitation of the two-magnon mode: femto-nanomagnonics

The use of magnons in dielectric materials for data transfer and processing devices has been recently discussed in view of the progresses achieved in magnonics research. It has been pointed out that magnons have the potential to bring the clock frequency of future magnon-based computational devices into the THz regime [41]. The high-frequency implies also short wavelength, which would allow the miniaturization of the technology. The optical stimulus represents a promising alternative to the more traditional schemes, involving a microwave resonant excitation and electrical detection of spin waves. However, the wavelength of the magnons excited in the aforementioned works lies in the micrometer scale and, as far as the frequency of the spin waves is concerned, the THz regime is barely achieved. Note that this is a direct

consequence of accessing magnons at the center of the Brillouin zone, which have the shortest wavevector and lowest frequency (see figure 2). This has been considered an intrinsic disadvantage of the optical approach, since the wavevector of light seems to restrict the excitation of spins to magnons with wavevector close to 0. More generally, the idea of a manipulation of the magnetic order on a sub-100 nm scale by means of visible electromagnetic radiation has been hitherto believed highly unlikely.

However, the magnon dispersion of a cubic antiferromagnet indicates that in principle such a limitation could be overcome by accessing spin excitations near the edges of the Brillouin zone. The dispersion curve can be calculated following a conventional procedure [22], considering the spin deviations (i.e. spin flips generating the spin waves or magnons) from the fully aligned antiferromagnetic ground state. Figure 2 reveals that high-wavevector spin waves for the

Heisenberg antiferromagnet KNiF_3 have periods on the femtosecond time-scale and wavelengths on the order of 1 nm.

Despite the absence of an ultrafast stimulus matching simultaneously the frequency and the wavevector of such magnons, it is possible to access this region of the dispersion via the Raman effect [15, 21, 51]. In our discussion we have hitherto neglected the contribution to equation (2) of the term containing two spin operators at different atomic sites. While this approximation is accurate in the description of the one-magnon physics [15, 21], it does not hold when we take into account the *two-magnon scattering* (2M), consisting in the creation or destruction of pairs of magnons. The Raman shift of the photon frequency is proportional to the sum $\omega_1(\mathbf{q}) + \omega_2(\mathbf{q}')$ of the frequencies of the two magnons. The total transferred wavevector is $\mathbf{k} = \mathbf{q} + \mathbf{q}'$. In principle, couples of magnons with wavevectors ranging over the whole Brillouin zone may contribute to this process. However, the experimental Raman spectra [15, 21] demonstrate that only the magnons with wavevectors near the edges of the Brillouin zone play an active role in the two-magnon process. Since in this region the dispersion flattens, the density of states is here dominant. Given the relative size of the wavevectors of light and of the Brillouin zone boundaries, it follows that $\mathbf{q} = -\mathbf{q}'$. Since two magnons are created with wavevectors equal in amplitude and opposite in direction, the symmetric dispersion relation plotted in figure 2 determines that the two frequencies of the magnons are equal $\omega_1(\mathbf{q}) = \omega_2(\mathbf{q}') = \omega(\mathbf{q})$.

A first approach to the explanation of the 2M mode could involve the extension of the one-magnon mechanism (i.e. spin-orbit coupling mediated spin-flip) to the second-order. The resulting cross section is several orders of magnitude smaller than in the case of the one-magnon scattering [15, 21]. This is in striking disagreement with the experimental observation that the two-magnon mode is of comparable, or even higher, intensity than the single-magnon mode [15, 21]. Therefore a different and more efficient scattering mechanism was proposed to explain this physical process, the so called exchange-scattering mechanism [15, 51]. In this framework, a light-induced modification of the exchange interaction perturbs the magnetic system in an equivalent way to two spin flip events, one on each sublattice, such that the total spin remains unchanged $\Delta S^z = 0$ (see figure 9). Theoretical frameworks addressing the photo-induced modification of the exchange interaction have been recently formulated [52–54]. Note that this argument explains also why this magnetic mode has never been observed in a ferromagnet, since in such a material flipping two spins gives rise to $\Delta S^z = \pm 2$.

Since the 2M is a Raman-active mode, an ultrashort laser pulse is indeed able to impulsively trigger it, provided that the requirements in terms of pulse duration and polarization are satisfied [55, 56]. However, the spin dynamics subsequent this photo-excitation is not trivial to detect and to interpret. Since the 2M process entails no modification of the total spin (i.e. $\Delta S^z = 0$), the transient magneto-optical Faraday and Kerr effects, which measure light-induced variations of the total spin, fail to track the dynamics of such a magnetic excitation. On the other hand, second-order magneto-optical

Figure 9. The 2M excitation is equivalent to a spin-flip event per sublattice. Thus, the magnetization of each sublattice ($S^\uparrow$ and $S^\downarrow$, represented by the two red arrows with opposite orientation) and, therefore, the antiferromagnetic vector (L^z , blue arrow) is decreased in the excited state. The z -direction indicates the axis along which the spins are aligned. The sum of the spins of the two sublattices, thus the total magnetization, vanishes both in the ground and in the excited state.

effects, which depend quadratically on the transversal spin components (i.e. on the spin deviations), are expected to succeed. A pair of spin flip events can thus be detected even if the total spin is unaffected [36]. Such a magneto-optical effect is a function of the correlation between spins belonging to different ionic sites, exactly like the Heisenberg exchange [22, 36]. In particular, the last term of equation (2) defines a magneto-optical effect proportional to the spin correlation function. Thus the spin dynamics induced by the 2M mode is tracked by measuring the time-evolution of the last term in equation (2). It was demonstrated that this is possible by detecting the transient rotation of the probe polarization [22]. Moreover, an analytical quantum mechanical model revealed that the spin correlation function has the same time-dependence of the longitudinal components of the antiferromagnetic vector $\mathbf{L}$ [22]. This result is consistent with the symmetry of the light-matter interaction energy describing the 2M-mode, which contains the spin correlation function [22]. The quadratic terms in the transversal spin components imply that the spin dynamics is symmetric in the x - y plane (given z the direction of the spins before the photo-excitation). Consequently, any macroscopic spin precession is forbidden, since a transversal component cannot be selected to initiate the precession. Typically the spin-orbit coupling is described by a term in the energy linear in the spin deviation (see the first two terms in equation (2)). Thus the generation of a macroscopic spin precession becomes in this case symmetry-allowed. On the other hand, the aforementioned argument shows that the spin dynamics triggered by the photo-generation of the 2M mode is exclusively longitudinal, which makes this regime unique.

An excellent system for the all-optical excitation and detection of the dynamics of high-frequency and shortest-

Figure 10. The transient rotation of the probe polarization was measured with the electric fields of the pump and the probe beams linearly polarized along the orthogonal directions. The pump fluence was set to $\approx 8.6 \text{ mJ cm}^{-2}$. The corresponding dynamics of L^z (blue arrows) is schematically represented. When the pump pulses impinge on the sample (0 delay) L^z decreases, due to the generation of magnons. At positive delays, oscillations at the frequency of the 2M mode are visible.

wavelength magnons is the cubic Heisenberg antiferromagnet KNiF_3 . In this material the Raman cross section of the 2M mode is so high that it dominates the whole spectrum [57]. A time-domain excitation of the 2M mode in KNiF_3 ($\nu_{2M} \approx 22 \text{ THz}$, period $\approx 45 \text{ fs}$, wave vector $\approx 10^7 \text{ cm}^{-1}$, wavelength $\approx 1 \text{ nm}$) can be achieved by the ISRS mechanism, provided that the duration of the stimulus is shorter than the period of the magnetic mode. A successful impulsive excitation of such high-frequency magnons therefore demands laser pulses significantly shorter than 40 fs. These requirements were met by using linearly polarized sub-20 fs laser pulses, with a central photon energy of 2.2 eV. For the probe, equally short pulses centered around 1.3 eV and with a polarization perpendicular to that of the pump were employed (see figure 4(b)).

The data in figure 10 show oscillations at the frequency of 22 THz and a lifetime of 500 fs. Although these values are in excellent agreement with the position and bandwidth of the 2M-peak observed in the Raman spectrum of KNiF_3 , a conclusive assignment of these oscillations requires further experimental evidence. The temperature dependence of the effect revealed that the frequency and the lifetime of the oscillations decrease as the Néel point is approached. This trend matches qualitatively and quantitatively the results obtained via spontaneous Raman spectroscopy [22]. Therefore it can be safely concluded that the signal displayed in figure 10 corresponds to the 2M-mode, entailing that coherent magnons with 45 fs period and wavelength of 1 nm were excited and probed [22, 58]. Given these numerical values, the collective spin-excitations investigated in this work were named *femto-nanomagnons*.

The phase and the amplitude of the short-wavelength magnons were coherently manipulated by changing the excitation conditions (see figure 11). More specifically, rotating the polarization of the pump beam by 90° results in

Figure 11. The blue and black dotted traces were obtained using a single-pump beam linearly polarized along the z axis (blue circles) and the x axis (black squares), as defined in figure 4(b). The phase of the oscillations is shifted by π . The single-pump measurement is shown again for the sake of comparison with the results of the double-pump experiments. The green-circle data set was observed by delaying two pump pulses by $T_{\text{pumps}} = 45 \text{ fs}$, that is, the period of the 2M mode. The coherent amplification of the signal is observed. The coherent oscillations were nearly quenched, when the delay between the two pump pulses was set to $T_{\text{pumps}} = 22.5 \text{ fs}$. The fluence of both pump beams was set to $\approx 1.5 \text{ mJ cm}^{-2}$. The temperature was 80 K. The probe beam was polarized along the x -axis.

the reversal of the sign of the signal, i.e. a π -shift of the phase of the magnons. Aiming at controlling also the amplitude of the oscillations, a double-pump excitation scheme was developed. When the two train of pump pulses were delayed by an amount of time equal to the period of the 2M mode, the amplitude of the oscillations was enhanced. On the other

hand, by setting the delay equal to a half-period of the 2M, the signal was quenched. Consequently a coherent manipulation of the order parameter in an antiferromagnet has been achieved on the 10 fs time-scale.

6. Femto-nanomagnonics: future perspectives

The understanding of the femtosecond spin dynamics triggered by exchange magnons with wavevector near the edges of the Brillouin zone is in its infancy.

The purely longitudinal nature of the oscillations of the order parameter suggests that the classical equations of motion for a magnetic system (Landau–Lifshitz type) cannot be employed. A comprehensive picture of this regime of spin dynamics, in which classical phenomenology and a full-quantum approach are compared, is still lacking.

Moreover, the space propagation properties of the 2M-mode has to be investigated. Since the spectrum of this magnetic excitation does not consist of a single mode but of a continuum [22, 57], it is not trivial to predict how the femto-nanomagnons propagate in space. Second- and third-order dispersion effects should be taken into account. Here, we point out that this issue is fundamentally engaging, considering recent results reported for thermal magnons with a wavelength of 100 nm in a dielectric magnetic material [59]. It was experimentally demonstrated that these quasiparticles diffuse on a length-scale which is orders of magnitude larger than the wavelength. Based on this evidence, it was argued that short-wavelength magnons behave more like particles than waves: even if the mean-free path is on the nanometer regime, they can propagate after millions of collisions on the micrometer scale [59].

The approach employed to trigger and detect spin excitations with nanometer wavelengths in KNiF_3 is very general and can be applied to several material classes. It is important to underline that the ISRS excitation of the two-magnon mode allows the manipulation of the spin system on the sub-wavelength regime without any dissipation. The photon energy of the pump beam was the same employed to disclose the zero-absorption regime in the same material [29]. The possibility to trigger coherent oscillations of the nearest-neighbors spin correlation function in the absence of dissipation may contribute to improve the present understanding of strongly correlated materials. In the framework of the Hubbard model such an excitation is supposed to initiate a coherent dynamics of the band-gap, which has not been observed yet. Moreover, it has been reported that the correlations among nearest-neighbor spins play a significant role in the coupling allowing superconductivity in high-Tc cuprates [60]. An experimental investigation of the coherent femto-nanomagnons in these materials may shed more light on this complicated and long-standing problem.

The femto-nanomagnonics regime offers thus unique possibilities to push the frontiers of fundamental knowledge in the field of light–matter interaction on the femtosecond time-scale. On top of that, it represents an intriguing concept for the development of magnon-based devices.

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