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Controlling high frequency spin-waves dynamics using double-pulse laser excitation

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Manipulating spin waves is highly required for the development of innovative data transport and processing technologies. Recently, the possibility of triggering high-frequency standing spin waves in magnetic insulators using femtosecond laser pulses was discovered, raising the question about how one can manipulate their dynamics. Here we explore this question by investigating the ultrafast magnetization and spin wave dynamics induced by double-pulse laser excitation. We demonstrate a suppression or enhancement of the amplitudes of the standing spin waves by precisely tuning the time delay between the two pulses. The results can be understood as the constructive or destructive interference of the spin-waves induced by the first and second laser pulses. Our findings open exciting perspectives towards generating singlemode standing spin waves that combine high frequency with large amplitude and low magnetic damping.

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I. INTRODUCTION

The last few decades have seen the emergence of a very promising paradigm for information transfer and processing: magnons, the quanta of spin waves (SWs), which exist in both conducting and insulating materials can be used to process information [1-7]. The interest in SWs is due to their large variety of useful properties at room temperature, such as low energy dissipation, long propagation distances up to millimeters, high propagation velocity of several km/s, broad high-frequency spectrum from GHz to THz, and tunable wavelength down to the nanoscale [1-8]. These properties may offer a faster and less energy-consuming alternative to concurrent charge-based semiconductor computing technology, which is associated with inevitable losses by current-induced Joule heating [7,9,10]. SWs also provide an opportunity to explore fascinating fundamental spin physics arising from the interplay of the dipolar and exchange interactions [8,11]. This is particularly manifest in the complex dispersion relations comprising magnetostatics and exchange SW modes, which can be strongly influenced by the thickness and lateral dimensions of the sample, the direction of the magnetization, and the strength of the applied external magnetic field [8,11].

Motivated by the above-mentioned potential for applications and rich physics, intense research activities are currently carried out to study and understand the generation, manipulation, and detection of SWs in several of the continuously developing fields of modern magnetism such as magnonics [1-5], spintronics [4,12,13], magnetoacoutics [14-17] and femtomagnetism [6,18-22]. Among all magnetic materials, yttrium iron garnet (Y₃Fe₅O₁₂, YIG) has attracted a lot of interest for data transport and processing technologies because it exhibits the lowest magnetic damping [23]. Several useful magnetostatic SWs devices have already been realized using YIG [24-27], however, due to its small saturation magnetization, their frequencies do not exceed a few GHz, which does not satisfy the high frequency requirement for ultrafast applications. In 2019, the possibility of triggering high frequency exchange standing SW (SSW) modes at very weak magnetic field was discovered in bismuth-substituted YIG (Bi-YIG) using femtosecond laser pulses [21,22]. The high frequency of the SSWs together with their low magnetic damping and the large magneto-optical responses of Bi-YIG are extremely helpful for ultrafast photomagnonic applications [21,22]. However, the SSWs are generated via mechanisms based on light absorption [21,22], which do not allow controlling the SSWs dynamics using the polarization of the pump pulses. Therefore, the fundamental question - how to control the SSWs dynamics in Bi-YIG - remains open.

In this work, we report on ultrafast magnetization and SSW dynamics in the nanoscale thin film of Bi-YIG. The sample has a large magneto-optical response and a high growth-induced inplane magnetic anisotropy allowing us to excite and probe high-frequency standing spin waves by performing femtosecond magneto-optical pump-probe experiments. Using an approach based on double-pulse excitation, we demonstrate an amplification or suppression of the spin wave resonance modes by precisely tuning the time delay between the two laser pulses. The results can be understood as the constructive and destructive interference of the spin waves induced by the first and second laser pulses.

II. SAMPLE PROPERTIES AND EXPERIMENTAL METHODS

The experiments are carried out in a 135-nm thick ferrimagnetic Bi₁Y₂Fe₅O₁₂ single-crystalline garnet film, grown by pulsed laser deposition (PLD) on a (100)-oriented gadolinium gallium garnet (Gd₃Ga₅O₁₂, GGG) substrate. More information on the sample growth condition and the PLD used can be found in Ref. [28,29]. The static MO and magnetic properties of the sample are studied using a custom-designed broadband MO spectrometer based on the 90°-polarization modulation technique [30,31]. The MO Faraday and Kerr spectra are in good agreement with previous studies of the MO properties of Bi-YIG. For instance, the MO Faraday rotation reaches large values of -3.84 °/µm at 2.45 eV and 28 °/µm at 3.26 eV, which is useful for photomagnetic devices. The in-plane saturation field (approximately 5 mT) is about 50 times smaller than the perpendicular saturation field (260 mT) [Figs. 1(a) and 1(b)]. The normalized remanence (M_r/M_s) is 0.85 and 0.02 for the in plane and perpendicular configuration, respectively. These results show a strong in plane magnetic anisotropy. This is due to an in-plane growth induced magnetic anisotropy constant $K_{\rm U} \sim -5750$ J/m³, which is very large compared to the monocrystalline cubic anisotropy constant $K_{\rm C} \sim -400$ J/m³. These parameters are determined from a micromagnetic simulation of the dependence of the ferromagnetic resonance mode frequency as a function of the external magnetic field. They are connected to the magnetic uniaxial and cubic anisotropy fields via $H_{\rm U} = \frac{2K_{\rm U}}{\mu_0 M_0}$ and $H_{\rm C} = \frac{2K_{\rm C}}{\mu_0 M_0}$, respectively. Using a saturation magnetization (Ms) in Bi₁Y₂Fe₅O₁₂ at room temperature of 135 kA/m, a uniaxial anisotropy field $\mu_0 H_U \sim -85$ mT and a cubic anisotropy field $\mu_0 H_C \sim -6$ mT are obtained.

The laser-induced ultrafast magnetization dynamics was investigated using the pump-probe configuration sketched in Fig. 1(c). An amplified laser system producing 120 fs pulses with a central photon energy of 1.55 eV and a repetition rate of 1 kHz is used as a light source. The output of the amplifier system was divided into two beams to generate the pump and probe

pulses. The probe pulse is frequency doubled to 3.1 eV and focused onto the sample at normal incidence in a spot dimeter of 60 µm. The pump pulse is kept at the fundamental of the amplifier at 1.55 eV and introduced into an optical delay line to control the delay t between the pump and probe. To perform the double excitation experiments, the pump is further divided in two equal parts. The delay Δt between them can be adjusted with a second optical delay line [see Fig. 1(c)]. The two pump pulses are recombined into two perfectly overlapping collinear pulses by a second beamsplitter and focused onto the sample under a small incidence angle of approximately 1° in a spot diameter of 250 µm. The probe is incident onto the sample from the substrate side. Due to the large gap energy of GGG [32], the probe traverses it and reaches the Bi-YIG layer. The reflected probe beam measures both the differential changes of the MO polar Kerr rotation $\Delta \Theta_{\rm K}(t)$ and reflectivity $\Delta R(t)$ induced by the pump pulses as a function of the pump and probe delay t using a polarization detection scheme. This configuration offers the advantage to detect the acoustic strain pulses generated in Bi:YIG via time-domain Brillouinscattering (TDBS) modulations of the reflectivity signal $\Delta R(t)$ while they propagate in GGG [16,17,21]. The external magnetic field H_{ext} is applied perpendicular to the film plane. All measurements are performed at room temperature.

III. RESULTS AND DISCCUSSION

To demonstrate the possibility of generating high frequency SSWs in our sample, we first investigate the ultrafast magnetization dynamics triggered by a single pulse excitation. Figure 2(a) shows the time-resolved magneto-optical Kerr effect (TR-MOKE) induced by a pump energy density $F_{pump} = 120 \text{ mJ cm}^{-2}$ for $H_{ext} = 0.14 \text{ mT}$. We chose this value for H_{ext} because it provides the highest induced spin precession amplitude. The TR-MOKE signal shows complex oscillations, which are formed by six well speared resonance modes in the range between 0.8 and 21 GHz, as seen in the fast Fourier transform (FFT) spectrum in the top inset of Fig. 2(a). The precession frequencies of these modes have a quadratic dependence on the mode number (Fig. 2(a), bottom inset). This variation can be well described by the dispersion relation of SSW, which is written, in the case of our experimental configuration, as $\omega(k) = \omega_0 + \gamma D_{ex}k^2$ [33,34], where ω is the angular precession frequency, ω_0 is the angular frequency of the fundamental mode which depends on H_{ext} and the effective magnetic anisotropy field, γ is the gyromagnetic ratio, D_{ex} is the exchange stiffness, and $k = n\pi/d$ represents the wave-vector of the standing spin waves given by d =135 nm. Indeed, a fit of the experimental data with the SSWs dispersion relation using $(\gamma/2\pi) = 28$ GHz/T yields an excellent agreement with the experimental results (Fig. 2(a), bottom inset). We obtain an exchange stiffness $D_{ex} = (5.9 \pm 0.2)$ 10⁻¹⁷ Tm² in good agreement with those reported in the literature for YIG [35-37] and Bi-YIG [21]. This clearly demonstrates the SSW nature of the observed magnetic resonance modes. We mention that the detected SSW amplitudes can be reproduced from theoretical point of view using a multilayer and transfer matrix model [38]. Such a full theoretical study goes however beyond the scope of the present paper.

The amplitude and phase of the SSWs modes do not depend on the linear polarization angle. This demonstrates that non-thermal mechanisms of the inverse Cotton-Mouton effect [39-41] and the photo induced magnetic anisotropy [42-45] are not at the origin of the excitation. The excitation of the SSWs can be related to an ultrafast change of magnetic anisotropy by incoherent and coherent phonons, as both mechanisms can induce a magnetization precession which is not sensitive to laser polarization [14-16,18,21,46-49]. The incoherent phonons change the magnetic anisotropy due to the temperature dependence of magnetic anisotropy constants [18,21,46-48], whereas the coherent phonons change the magnetic anisotropy via inverse magnetostriction [14-16,21,49]. The generation of incoherent phonons, i.e. heat, should also induce demagnetization dynamics due to the transfer of the heat energy from the hot phonons to the cold spin system via phonon-magnon coupling. To confirm this possibility, we measured the TR-MOKE induced for a saturation magnetic field of $H_{ext} = 0.34$ T [Fig. 2(b)]. A subpicosecond decrease of the magnetic order is observed, suggesting a phonon-magnon coupling ≤ 0.6 ps. The remagnetization dynamics shows an extremely long-time scale, which is in good agreement with the poor thermal conductivity of iron garnets [50-52]. On the other hand, the generation of coherent phonons is revealed by the TDBS observed in the ΔR signal TDBS frequency exactly equals the value $f_{GGG} =$ [Fig. 2(c)].Indeed, the $2E_{\rm p}V_{\rm GGG}^{\rm L}\sqrt{n^2-\sin^2\theta}/1240 = 64$ GHz, calculated from the probe photon energy $E_{\rm p} = 3.1$ eV, the longitudinal sound velocity $V_{GGG}^{L} = 6400 \text{ m/s}$ of GGG [53], its refractive index $n \approx 2$ [32], and the angle of incidence $\theta \approx 0^\circ$. The generation of the incoherent and coherent phonons is related to the absorption of the pump by one and two photon absorption (TPA) processes. The TPA process is revealed by the quadratic dependence of the demagnetization and reflectivity dynamics as a function of F_{pump} , as seen in the insets of Figs. 2(b) and 2(c). The one photon absorption is related to the excitation of the phonon assisted ${}^{6}S \rightarrow {}^{4}G$ electronic d-d transitions of Fe³⁺ ions in octahedral sites, which have a small amplitude, and therefore play a minor role at high F_{pump} , where TPA is dominating [21,54]. We note that these electronic states have a very short lifetime and transfer the heat energy to the lattice within hundreds of femtoseconds via electron-phonon coupling [51].

To explore the ultrafast manipulation of SSW dynamics we use an approach based on double pulse excitation. Figure 3(a) shows the TR-MOKE signal induced by single- and double-laser excitation with a separation time Δt slightly different from 0 in order to avoid interference between the pump beams. We choose $\Delta t = 1$ ps which is much shorter than the 50 ps precession period of the fastest mode. The laser fluence of each pump beam is $F_{pump} = 120 \text{ mJ cm}^{-2}$ and $H_{\text{ext}} = 0.14$ T. The TR-MOKE induced by double-laser excitation shows a magnetization precession with an amplitude larger than the one induced by a single pulse excitation. This is particularly interesting as, for a single pump beam, the fluence of 240 mJ cm⁻² is higher than the sample's damage threshold (145 mJ cm⁻² evidenced by a dramatic decrease of the static MO response of the sample) and therefore cannot be used to excite the SSW dynamics. The enhancement of the SSW amplitude can be explained by a constructive interference of the spinwaves induced by the first and second laser pulses. We anticipate here that by using a sequence of more than two laser pulses with $\Delta t \sim 0$ can one further and further increase the SSW amplitude. From a technological point of view, this enhancement of SW amplitudes should facilitate their detection, which can improve the performance of SWs-based data transport and processing technologies.

To investigate the possibility of suppressing SSWs, we measured the TR-MOKE induced by double pulse excitation with $\Delta t = 100$ ps [Fig. 3(b)]. This separation time corresponds to the half of period of the SSW with mode number n = 2, which has the largest amplitude. A clear difference is observed between the TR-MOKE data measured for $\Delta t = 100$ ps and $\Delta t \sim 0$: the dominant SSW mode for the measurement with $\Delta t = 100$ ps has a period about 2 times larger than the dominant high-frequency mode for $\Delta t \sim 0$. To better highlight this difference, we show in the inset of Fig. 3(b) the corresponding FFT spectra, which clearly reveal a suppression of the SSW with mode number n = 2. This suppression can be related to a destructive interference of the SSW with n = 2, as the delay between the two pulses is equal to the half of its period.

To further investigate the suppression of SSW modes, we measured the TR-MOKE induced by double pulse excitation with $\Delta t = 220$ ps and $\Delta t = 56$ ps, which correspond to the half of period of the SSWs with the mode number n = 3 and n = 1, respectively [Fig. 4]. The FFT analyses of the TR-MOKE data clearly demonstrate that the SSW with mode number n = 1 is totally suppressed for $\Delta t = 220$ ps, while the SSW with mode number n = 3 is completely suppressed for $\Delta t = 56$ ps [insets of Figs. 4(a) and 4(b)]. This supports the conclusion that the amplification or suppression of SSWs is related to a constructive or destructive interference of the spin-waves induced by the first and second laser pulse. We note that this approach can in principle be used

to control propagating spin waves, which is also of great interest from both fundamental science and technological applications [1-5,7,23]. On the other hand, let us mention here that an approach based on a train of fs-laser pulses with a high repetition rate of 1 GHz has been recently used for significant enhancement of the amplitude and narrowing the spectrum for propagating SWs [20,55-57]. Our results predict that using the same approach with a high repetition rate resonant to a SSW mode should similarly allow a very large increase of its amplitude, which can open perspectives towards the generation of large amplitude single SSW mode.

IV. CONCLUSION

In summary, we investigated the ultrafast magnetization and SSW dynamics in a nanoscale film of Bi-substituted iron garnet. The sample has a large MO effect and low magnetic damping, which ideally fulfills the conditions of photo-magnonics applications. We demonstrate that femtosecond laser excitation allows triggering high frequency SSW modes and probing their dynamics via the time resolved magneto-optical Kerr effect. Using an approach based on double-pulse excitation, we demonstrate an amplification or suppression of desired spin-wave resonance modes by precisely tuning the time delay between the pulses. The results can be understood by considering constructive or destructive interference of the SWs induced by the first and second laser pulses. These findings open exciting perspectives towards the generation of single mode SSWs that combine high frequency with large amplitude and low magnetic damping.

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Fig. 1. Static magnetic properties of the garnet sample and experimental TR-MOKE configuration. (a, b) Normalized (a) polar and (b) longitudinal Kerr magnetization hysteresis loops. The inset shows a magnified view of the region around 0 mT. (c) Sketch of the double-pump experiment: a sequence of two collinear pulses with a central photon energy of 1.55 eV are focused onto the sample under a small incidence angle of approximately 1° in a spot diameter of 250 μ m. The separation time Δt between the two pump pulses is controlled using an optical time delay. The probe pulse at a central photon energy of 3.1 eV probes the Bi:YIG film at normal incidence from the backside through the GGG substrate.



Fig. 2. Ultrafast magnetization and reflectivity dynamics in the Bi₁Y₂Fe₅O₁₂ film. (a), (b) $\Delta \Theta_{\rm K}(t)/\Theta_{\rm Ksat}$ induced by $F_{\rm pump} = 120$ mJ cm⁻² for (a) $H_{\rm ext} = 0.14$ T and (b) $H_{\rm ext} = 0.34$ T. (c) ΔR induced by $F_{\rm pump} = 120$ mJ cm⁻². Insets (a): (top) FFT spectrum of $\Delta \Theta_{\rm K}/\Theta_{\rm Ksat}$ data. (Bottom) Spin precession frequency as a function of the mode number. The solid line is the fit obtained using the SSW dispersion relation ω (k) = $\omega_0 + \gamma D_{ex}k^2$. Inset (b): (top) $\Delta \Theta_{\rm K}(t =$ 85 ps)/ $\Theta_{\rm Ksat}$ as a function of $F_{\rm pump}$. The solid line is the fit to the data using $\Delta \Theta_{\rm K}/\Theta_{\rm Ksat} =$ $a (F_{\rm pump})^2$. (Bottom) $\Delta \Theta_{\rm K}(t)/\Theta_{\rm Ksat}$ around t = 0. Insets (c): (top) FFT spectrum of ΔR data. (bottom) $\Delta R(t = 85$ ps) as a function of $F_{\rm pump}$. The solid line is the fit to the data using $\Delta R =$ $b (F_{\rm pump})^2$.



Fig. 3. Enhancement and suppression of spin waves by double-laser excitation. (a) $\Delta \Theta_K(t) / \Theta_{Ksat}$ induced by one- and double-pulse excitation with $\Delta t \sim 0$. (b) $\Delta \Theta_K(t) / \Theta_{Ksat}$ induced by a double-pulse with separation times $\Delta t \sim 0$ and $\Delta t = 100$ ps. For better visualization a vertical offset is introduced. For each pump beam the laser fluence is 120 mJ cm⁻². The inset shows the FFT spectrum of the $\Delta \Theta_K / \Theta_{Ksat}$ data.



Fig. 4. Manipulating high frequency spin-waves dynamics using double-laser excitation. $\Delta \Theta_{\rm K}/\Theta_{\rm Ksat}$ induced by double-laser excitation with $\Delta t \sim 0$ and $\Delta t = 220$ ps. (b) $\Delta \Theta_{\rm K}/\Theta_{\rm Ksat}$ induced by double-laser excitation with $\Delta t \sim 0$ and $\Delta t = 56$ ps. The inset shows the FFT spectra of the $\Delta \Theta_{\rm K}/\Theta_{\rm Ksat}$ data. For better visualization a vertical offset is introduced in $\Delta \Theta_{\rm K}/\Theta_{\rm Ksat}$ data and the FFT spectrum.