Stimulated $k$-vector selective magnon emission in NiFe films using femtosecond laser pulse trains

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We demonstrate stimulated coherent emission of localized and propagating spin waves in 20 nm thick NiFe films using a 1 GHz repetition rate femtosecond laser. As the laser pulse separation is shorter than the time required for the excited magnons to dissipate, magnons having a frequency equal to a multiple of 1 GHz are coherently amplified as each new pulse adds magnons predominantly in-phase with the existing population. Using scanning micro-Brillouin Light Scattering (BLS) we observe this coherent amplification as strong 1 GHz spaced peaks in the BLS spectrum. In contrast to single pulse rapid demagnetization, where the spin wave amplitude grows linearly with laser power, we observe an exponential power dependence, clearly indicating the stimulated nature of the magnon emission. By tuning the applied magnetic field, we can select which wave vectors to coherently amplify. This is directly confirmed using spatial mapping of the generated magnon population, which shows that both localized and propagating spin waves can be excited in the stimulated regime and
that the direction of the propagating spin waves can be controlled by the in-plane component of the applied field. Our results enable a new platform for photo-magnonics where sustained coherent spin waves can be utilized.

Magnonics has emerged as a central research topic in nanomagnetism, with rich physics and an increasing number of novel phenomena thanks to the unique field-tunable properties of spin waves (SWs) and a wide range of metallic and insulating magnetic materials. As the wavelength of SWs can be several orders of magnitude smaller than its electromagnetic radiation counterparts at the same frequency, the possibility of scaling down high-frequency devices using magnonics offers excellent prospects for miniaturization.

SWs can be excited using a wide range of mechanisms. While the most straightforward and conventional SW generation mechanism is that of an externally applied microwave field using RF antennas, the more recent spin transfer-torque and spin Hall effects generated by direct currents through nanodevices have made it possible to generate truly short wavelength, highly non-linear, and very high intensity SWs on the nanoscale. SWs can also be generated optically using focused femtosecond laser (fs-laser) pulses inducing rapid demagnetization of the local magnetization. Single-pulse excitation schemes, where the system relaxes into equilibrium before the arrival of the second pulse, have been studied extensively in metals and dielectrics. As the pump pulses are usually separated by about 12 ns or more, corresponding to a repetition rate of 80 MHz or lower, and the SW decay time in typical ferromagnetic metals is a few nanoseconds, SWs excited from an individual pulse will have damped out before the arrival
of the next pulse.

While most all-optical studies conducted so far have used stroboscopic pump-probe techniques such as time resolved Magnetooptic Kerr effect (TR-MOKE) or Faraday effect to study the immediate SW response after each individual pulse, time-averaged techniques such as Brillouin Light Scattering (BLS) microscopy have been inadequate due to the low duty cycle. Most TR-MOKE experiments yield a highly resolved temporal picture of the coherent evolution of the magnetic state but cannot detect incoherent (e.g. thermal) magnons, nor other excitations such as phonons. BLS microscopy, on the other hand, is an extremely versatile and sensitive technique, with a very wide frequency range, good spectral resolution, simultaneous detection of coherent and incoherent dynamics, and also offering high wave-vector resolution. As an added benefit, it also allows for simultaneous detection of both magnetic and non-magnetic excitations (such as phonons) with the same frequency and spatial resolution. It would therefore be highly advantageous to use BLS microscopy for the study of fs-laser induced SW excitations, which then requires a higher duty cycle, i.e. much shorter time in-between laser pulses.

The first steps towards reducing the time between consecutive pulses were taken by employing pump-probe techniques with dual pump pulses. By tailoring the time delay between two such pump pulses, the precession in both single- and multi-layer systems could either be quenched or amplified. While carefully controlling the phase relation hence allowed for detailed selectivity of which SWs to excite and further amplify, the overall duty cycle was however not improved.

An experimentally much simpler approach is instead to increase the fs-laser repetition rate to
approach the time scale of the SW decay. As frequency comb based fs-lasers with GHz repetition rates have recently become commercially available, the very first studies of high repetition rate SW excitations in thick extended YIG films were reported. Using the inverse Faraday effect for excitation with a 10 μm laser spot size, and a conventional TR-MOKE pump-probe technique for detection, the authors were able to demonstrate coherently amplified excitation of SWs whose phase relation matched the time between consecutive pulses.

These pioneering works raise a number of intriguing questions. First, can high repetition rate fs-lasers also be efficiently used to excite and coherently amplify SWs in metallic thin films and devices using rapid demagnetization? Secondly, can the high-repetition rate lasers enable the use of time-averaged techniques such as BLS microscopy in the study of ultrafast magnetization dynamics? Finally, if the pump laser and the BLS laser are focused down to their diffraction limits, can both localized and propagating SWs with both small and large wave vectors be excited, detected, and controlled?

Here we attempt to answer these questions. Using a unique Brillouin light scattering (BLS) microscope, where we combine a diffraction limited BLS SW detection scheme with a diffraction limited high repetition rate (1 GHz) fs-laser inducing rapid demagnetization, we demonstrate stimulated and wave-vector selective emission of SWs at multiple harmonics of the fs-laser repetition rate. The high sensitivity of our BLS microscope resolves both localized and propagating SWs when the separation between the two laser spots are scanned to reveal the spatial profiles of the SWs. By tuning the magnitude and direction of the applied magnetic field, we can choose a specific
wave vector to be amplified coherently, and also steer the direction of the SW propagation. Our results clearly demonstrate that it is possible to excite and control SWs of high amplitude using a high repetition rate fs-laser and detect them with high sensitivity using BLS microscopy.

Figure 1: Schematic of experiment and experimental setup. a. The sample is pumped with a red (816 nm) 120 fs, 1 GHz-pulsed laser and probed by a continuous green laser (532 nm); the relative distance between the pump and the probe can be scanned over ±40 µm. The magnetic field $H_{\text{ext}}$ is applied at an oblique out-of-plane angle of 82°. b. The optical setup of the pump-probe experiment: BS – 50:50 Beam Splitter, PBS – Polarizing Beam Splitter. The sample is placed right below a 100x objective with NA = 0.75 to achieve diffraction limited focusing. The back-scattered beam is analysed using a 6-pass Tandem Fabry-Pérot Interferometer (TFPI) and detected with a single channel avalanche photodiode (APD).

Our pump-probe experiment is shown schematically in Fig. 1a: A pulsed laser with a repetition rate of 1 GHz, pulse length of 120 fs, and a wavelength of 816 nm is used as the pump,
whereas a continuous wave 532 nm laser is used as the probe. Inelastically back-scattered photons from the sample are analyzed using a Tandem Fabry-Perot Interferometer (TFPI) equipped with an avalanche photodiode (APD) in order to reveal information about the magneto-dynamics of the sample (Fig 1b). The experiment was performed at room temperature on a 20 nm thick polycrystalline permalloy (Py) thin film deposited on a sapphire substrate with negligible absorption at 532 nm and excellent heat conductivity. The permalloy film was coated with 2 nm of dielectric SiOx.

Fig. 2a shows the typical field-dependence of the thermal SW spectrum for the Py thin film sample when the pump laser is off. The sharp lower cut-off of the SW band can be clearly seen as it follows a Kittel-like frequency-vs-field dependence. A more gradual decay of the BLS counts are observed at higher SW frequencies as the SW wave-vector approaches the resolution limit of the BLS.
Figure 2: **Thermal and stimulated spin wave excitation in a 20 nm NiFe thin film.**

- **a.** Thermal SW spectrum vs. magnetic field.
- **b.** SW spectrum at a 1.8 mJ/cm² fs-laser pump fluence.
- **c.** BLS counts vs. frequency in a field of 600 mT for five different pump powers showing pronounced peaks at the harmonics of the 1 GHz repetition rate. **Inset:** BLS counts at 8 and 9 GHz vs. pump fluence, showing an exponential behavior.
- **d.** Power dependence of the BLS counts at the pump spot.
- **e.** Same when the probe is 1 µm away from the pump, showing SW propagation at 8 and 9 GHz.
Fig. 2b shows the corresponding spectrum at the same location when the fs-laser is turned on with an intermediate power of 10 mW, (pump fluence of 1.8 mJ/cm²). The spectrum changes completely and instead of being largely featureless, a number of distinct peaks appear at the harmonics of the 1 GHz repetition rate, with the strongest peaks having about an order of magnitude higher intensity than the thermal background. While the peaks seem to fall close to the bottom of the original SW band, there is now also sizable BLS counts well below the gap at these harmonics.

To better discern all features of the laser induced SW intensity, we show in Fig. 2c a detailed plot of the BLS counts vs. frequency at a field magnitude of 600 mT for four different laser powers and the thermal SWs. We first note that 5 mW seems to be the approximate threshold for any noticeable additional SW intensity compared to the thermal SWs. At this lowest power level, we can observe additional BLS counts at 7, 8, and 9 GHz, but not at any other harmonics. At the highest power levels, pronounced SW peaks are observed at all harmonics, although these three frequencies remain the most strongly affected. It is noteworthy that the BLS counts at 7, 8, and 9 GHz all show an exponential dependence on laser power. This can be seen in detail in the inset of Fig. 2c, where the counts for the FMR peak shows a strong exponential increase over the full range of laser fluences, while the 9 GHz peak shows two slopes with a weaker exponential growth above 3 mJ/cm². This exponential dependence is in stark contrast to all earlier studies of rapid demagnetization where the power dependence was essentially always linear. This observation is a clear indication that the excitation mechanism is stimulated in nature.

A number of qualitatively different frequency regions can be observed based on the data
taken at high laser power. At low frequency we find that the BLS counts decrease exponentially with harmonic number 2–4, which is again consistent with a stimulated excitation mechanism. At intermediate harmonic numbers (5–8) the BLS counts increase rapidly and reach a strong maximum at the FMR frequency of 8 GHz. For yet higher harmonics, the counts again fall, most precipitously between the 9th and the 10th harmonic, above which the impact of the fs-laser remains limited compared to the thermal SWs.

To complement the data shown for overlapping pump and probe lasers in Fig. 2c, we show in Fig. 2d&e a comparison of the power dependence of the peaks at both overlapping and separated laser spots, again for a field of 600 mT. While again all harmonics can be observed in the overlapping case, the number of peaks is dramatically reduced when the pump and probe spots are separated by 1 μm, with essentially only the 8 and 9 GHz peaks surviving. We are hence lead to conclude that only at these two frequencies do the laser pulses generate propagating SWs of any magnitude; the magnetodynamics at all other harmonics are consequently of a local nature.

To study the propagation characteristics of these SWs we measured the spatial extension of the spectra at a 600 mT applied field. Fig. 3(a) shows a lateral scan of the BLS counts relative to the pump spot along the direction perpendicular to the in-plane component of the applied magnetic field.
Figure 3: **Spatial profiles of the stimulated SW excitation.**

a. SW intensity (log scale) vs. relative distance from the fs-laser pump spot along the axis of SW propagation. b. BLS counts at 8 GHz as a function of the relative distance between the pump and probe beams. $\xi$ is the spin wave propagation length calculated from an exponential fit to all the data. c-f. 2D mode profiles for the four major excited harmonics (7, 8, 9 and 10 GHz). The modes corresponding to 8 and 9 GHz show propagation characteristics along the vertical axis while the modes at 7 and 10 GHz are localized at the pump spot.

The color plot (Fig. 3a) shows SW intensity at 8 GHz up to a distance 4 $\mu$m away on either side of the excitation spot. The mode at 9 GHz propagates up to 3 $\mu$m on either side. In contrast, the modes below 8 GHz and above 9 GHz are localized to the pump region. The maximum intensity of the peaks in the spectra as a function of the vertical coordinate is obtained by fitting the spectra.
to a Lorentzian function and is shown for the frequency 8 GHz (at 600 mT) for different pump powers (Fig.3b). By fitting the profiles to an exponential decay function of the form $\exp(-2x/\xi)$, where $\xi$ is the SW propagation length, we obtain a value $\xi = 1.85 \mu m$.

Full 2D spatial profiles of the SW modes are then obtained by raster scanning over the sample. Fig.3c-f. are the respective mode profiles of the 7, 8, 9 and 10 GHz peaks. The mode at 7 GHz is clearly localized at the pump spot. At 8 GHz the pump harmonic coincides approximately with the broadband ferromagnetic resonance peak and at 9 GHz with higher wave vector SWs. High amplitude SWs are then excited from the center and propagate in the direction perpendicular to the in-plane component of the applied field. Additionally, we find that it is also possible to steer the propagation direction by rotating the in-plane-component of the applied magnetic field. At 10 GHz, we do not find any measurable evidence for SW propagation. As SW frequencies of 10 GHz in a field of 0.6 T correspond to wave vectors just above the cut-off of the resolution of our BLS (see Fig.2a) we cannot say for certain that no SW propagation takes place at these high wave vectors. However, as the wave-vector cut-off for excitation and detection should depend similarly on spot size, and the spot size of the fs-laser ($\sim 800$ nm) is larger than that of the BLS laser ($\sim 350$ nm), this is unlikely.

To gain further insight into the stimulated nature of the SW emission, we performed micromagnetic simulations at an applied external field value of 0.6 T. The results were found to closely reproduce the overall behaviour of the experimental spectra and the spatial profiles of the experimentally observed SW modes. Fig.4 shows the spectrum of the harmonics excited at the pump spot
(a) and a distance 1 µm away (b). The agreement with the experimental data in Fig.2 is quite remarkable. While the magnetization dynamics at the laser spot is dominated by the FMR response, all other harmonics of 1 GHz are clearly present. At low frequencies, the amplitude decreases with harmonic number towards a shallow minimum at \( n = 4–5 \) after which the amplitude again increases towards the FMR response; at frequencies above FMR the amplitude at the laser spot again decreases exponentially with \( n \). As in the experiment, the spectral response is dramatically different 1 µm away from the laser spot, where the FMR peak and the first propagating SW mode dominate entirely and all other peaks are strongly suppressed.

Figure 4: Micromagnetic simulations. Simulated spectral density of the magnetization dynamics: a. at the pump location, and b. at a distance of 1 µm from the pump location. Insets show the instantaneous temporal evolution of the magnetization. The simulated amplitude (c, d, e, f) as well
as the phase profiles \(g, h, i, j\) for the strongest harmonics at 7, 8, 9, and 10 GHz are shown in the lower subfigures.

The insets show time traces of the \(x\) component of the magnetization at the two locations. While both time traces show a continuous SW intensity, the dynamics at the laser spot is more strongly affected by each pulse, whereas the response at 1 \(\mu\)m away varies smoothly and exhibits beating from the sum of the two dominant spectral lines.

Fig. 4c-h shows the spatial maps of the SW amplitudes and phase at modes corresponding to 7, 8, 9 and 10 GHz. As observed in the experiment, the SWs at 7 GHz correspond to a localized mode, as its frequency falls below the FMR. All higher frequencies correspond to propagating spin waves, of the Damon-Eschbach type, with wave-vectors perpendicular to the in-plane component of the applied magnetic field.

We believe that our demonstration of using a high repetition rate femtosecond laser for the stimulated excitation of continuous spin waves, with detailed control of the wave vector and probed using Brillouin Light Scattering microscopy, will enable a wide range of additional photomagnonic studies of both magnetic thin films and magnonic and spintronic devices. While our study used a single excitation spot, it will be straightforward to extend to multiple spots and/or shape the SW excitation region at will using optical techniques such as spatial light modulation. The possibility of adding stimulated SW excitation to the studies of STT and SHE driven devices is particularly intriguing. Outside of these immediate applications, using high repetition rate fem-
tosecond lasers for THz radiation emission in ferromagnet/heavy metal bilayers will likely also have a direct beneficial impact, as will much faster measurements of all-optical magnetic switching.

**Methods**

**Fs-laser pump, µ-BLS probe:** The pump beam was produced by a commercial Ti:Sa mode-locked laser with a 1 GHz repetition rate at a wavelength of 816 nm, a 30 fs Fourier-limited pulse duration and pulse energies up to 1nJ. The laser pulse stretched during propagation in the optical system and reached the sample with a duration of 120 fs. The laser was focused close to the diffraction limit by a 100x microscope objective with a N.A. of 0.75. The magneto-dynamics was probed using BLS microscopy, which has been described in detail elsewhere. A single-frequency continuous wave laser at 532 nm was focused to a near diffraction limited spot on the sample through the same objective as used for the pump beam. The probe beam undergoes a frequency shift due to scattering from spin waves. Back scattered light was collected and filtered using a polarizer and analyzed in a 6-pass Tandem Fabry-Perot Interferometer (TFPI) and detected using a single channel avalanche photodiode. The scattered light carries the phase, frequency and wavevector information from the spin wave. The spatial resolution is obtained at the expense of wavevector resolution by using the high-resolution microscope objective which forms a tight focus of the probe beam. The optical system is equipped with a pair of galvo-mirrors and lenses that allows the pump beam to be scanned over the sample and hence change the lateral distance between the pump and the probe beams.
Simulations  The micromagnetic simulations were performed on a 5.12*5.12 µm² permalloy film of 20 nm thickness using Mumax³. Periodic boundary conditions were used to suppress finite size effects expected for this sample. A saturation magnetization $M_s$ of 781.75 kA/m extracted experimentally from thermally excited FMR, exchange stiffness $A$ of 11.3 pJ/m and damping of 0.01 are the parameters used in the simulations. As in the experiment, an external field was applied at an oblique angle of 82°. To mimic the optical-pump effect spot used in the experiment, an instantaneous reduction of the saturation magnetization followed by a slower recovery where the magnetization relaxes back to its original value, applied at clock frequency of 1 ns in form of subtracting or adding the demagnetization tensor corresponding to the magnetization state at both the demagnetized and the recovered states. The pump-beam, as in the experiment, had a Gaussian profile with FWHM of 800 nm.

Data Availability Statement  The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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Competing Interests  The authors declare that they have no competing financial interests.

Authors contributions  A.A.A., S.M., A.A. and J.Å. designed the experiment. S.M. fabricated the samples. A.A. designed and assembled the optics related to the fs-laser, with assistance from D.L. and D.H., and characterized the optical parameters. S.M. and A.A.A. conducted most of the measurements and analysis. A.A.A. performed magnetic simulations with support from R.K. and M.D. who assisted with their
theoretical expertise. All authors contributed to the data analysis and co-wrote the manuscript.

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**Supplementary materials**

The directionality of the spin wave emission is verified by rotating the in-plane component of the applied field and measuring an area profile of the spin wave. We rotated the in-plane component by $30^\circ$ and the area maps also rotate by the same angle depicting an emission perpendicular to the in-plane component. This corresponds to a Damon-Escbach mode.
Figure S1: **2D area profiles at a different in-plane field angle.** The area maps of the modes at 7, 8, 9 and 10 GHz are shown in **a, b, c** and **d** respectively. **a.** shows the angle of the in-plane field component relative to the axis of the color map.

In the Fig[S1]a. and d., the modes are localized to the excitation spot whereas Fig[S1]b. and c. are propagating in the direction perpendicular to the in-plane field component.