

THz frequency combs from interfacial spatiotemporal overlap in spintronic multilayers

Hong-Guang Piao

hgpio@ybu.edu.cn

Yanbian University <https://orcid.org/0000-0001-5948-4327>

Je-Ho Shim

Yanbian University <https://orcid.org/0000-0002-2004-9798>

Yunxiu Zhao

Westlake University

Qoimatul Mustaghfiroh

Chungbuk National University <https://orcid.org/0000-0003-0841-8949>

Megha Jain

Chungbuk National University

Jihyeon Hwang

Chungbuk National University

Palwinder Singh

Chungbuk National University

Das Priyanka

Chungbuk National University

Kyungwan Kim

Chungbuk National University

Hee Jun Shin

Pohang Accelerator Laboratory, POSTECH

Jaehun Park

Pohang Accelerator Laboratory

Xiao You

Westlake University

Shilong Xiong

Chinese Academy of Sciences

Cai-Hua Wan

Chinese Academy of Sciences <https://orcid.org/0000-0002-5456-1052>

Xiufeng Han

Institute of Physics, Chinese Academy of Sciences <https://orcid.org/0000-0001-8053-793X>

Dong-Hyun Kim

Physical Sciences - Article

Keywords:

Posted Date: May 6th, 2026

DOI: <https://doi.org/10.21203/rs.3.rs-9613109/v1>

License: © ⓘ This work is licensed under a Creative Commons Attribution 4.0 International License.
[Read Full License](#)

Additional Declarations: There is **NO** Competing Interest.

THz frequency combs from interfacial spatiotemporal overlap in spintronic multilayers

Je-Ho Shim^{1,2*}, Yunxiu Zhao^{3,4*}, Qoimatul Mustaghfiroh³, Megha Jain³, Jihyeon Hwang³, Palwinder Singh³, Das Priyanka³, Kyungwan Kim^{3†}, Hee Jun Shin⁵, Jaehun Park⁵, Xiao You⁴, Shilong Xiong⁶, Caihua Wan^{6†}, Hong-Guang Piao^{1,2†}, Xiufeng Han⁶, and Dong-Hyun Kim^{2,3†}

¹*Department of Physics, College of Science, Yanbian University, Yanji 133002, China*

²*Institute of Quantum Science and Technology, Yanbian University, Yanji 133002, China*

³*Department of Physics, Chungbuk National University, Cheongju 28644, South Korea*

⁴*School of Engineering, Westlake University, Hangzhou 310024, China*

⁵*Pohang Accelerator Laboratory, POSTECH, Pohang 37673, South Korea*

⁶*Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, University of Chinese Academy of Sciences, Chinese Academy of Sciences, Beijing 100190, China*

* Je-Ho Shim and Yunxiu Zhao contributed equally to this work.

† **Corresponding author Emails:**

hgpiao@ybu.edu.cn (Hong-Guang Piao),

donghyun@cbnu.ac.kr (Dong-Hyun Kim),

wancaihua@iphy.ac.cn (Caihua Wan),

kyungwan@chungbuk.ac.kr (Kyungwan Kim),

Abstract

The terahertz (THz) spectral region bridges electronic and photonic domains, yet the development of compact, coherent, and tunable THz sources remains a central challenge. Spintronic emitters based on ultrafast spin dynamics have emerged as promising broadband THz sources. However, they have been studied primarily within linear spin–charge conversion frameworks, with nonlinear coupling phenomena, such as frequency comb generation, largely unexplored. Accessing this nonlinear regime would expand spintronic THz emitters beyond broadband transient emission, enabling coherent spectral structuring through ultrafast spin dynamics. Here, we report the emergence of nonlinear THz frequency combs in nano-engineered magnetic multilayers based on $[\text{Pt}/\text{Co}/\text{MgO}]_n$ and $[\text{Pt}/\text{Co}/\text{W}/\text{MgO}]_n$ heterostructures driven by femtosecond optical excitation. Using optical-pump/THz-emission time-domain stroboscopy, we resolve periodic spectral structures extending up to 15 THz. The comb formation originates from nonlinear coupling between two primary interfacial spin-dynamic modes: one dominated by the inverse spin Hall effect (~ 0.8 THz) and the other with ultrafast exchange-driven dynamics (~ 2.0 THz). We reveal that the key mechanism governing comb formation is the spatiotemporal overlap of these interfacial modes, in which sub-picosecond temporal coincidence and nanoscale spatial confinement enable efficient nonlinear interaction. This overlap defines a dominant interaction channel, giving rise to coherent higher-order spectral components that follow a universal comb relation. Our results establish a physically transparent framework for nonlinear THz generation in spintronic systems and demonstrate that interfacial ultrafast dynamics can be engineered to access frequency comb regimes, opening new opportunities for high-precision spintronic metrology and nonlinear THz spectroscopy of non-equilibrium spin phenomena.

Introduction

The terahertz (THz) spectral range, located between the microwave and infrared regimes, provides direct access to ultrafast electron, lattice, and spin dynamics on sub-picosecond timescales. Despite their broad potential applications in communication, imaging, and spectroscopy, the development of compact and coherent THz sources remains a long-standing challenge, commonly referred to as the “THz gap”¹⁻⁴.

Spintronic THz emitters based on magnetic heterostructures have recently attracted considerable attention as efficient broadband sources. In these systems, femtosecond optical excitation generates ultrafast spin currents and transient magnetization dynamics⁵⁻⁷, which are subsequently converted into charge currents through mechanisms such as the inverse spin Hall effect (ISHE)⁸⁻¹⁴, inverse Rashba–Edelstein effect¹⁴⁻¹⁷, inverse spin–orbit torque^{18,19}, and related interfacial spin–charge conversion processes. These advances, together with anomalous-Hall-effect-related ultrafast charge-current generation in magnetic materials²⁰⁻²³, have established spintronic emitters as a promising platform for THz generation⁸⁻²³.

However, most studies to date have described THz emission from magnetic heterostructures within a linear-response framework. The contributions from ultrafast demagnetization, spin transport, and spin–orbit conversion are typically treated as independent processes, often described within superdiffusive spin-transport and linear spin–charge conversion frameworks²⁴, while possible nonlinear interactions between coexisting spin-dynamic channels have received little attention. As a result, the accessible spectral range remains effectively limited, leaving much of the THz bandwidth unexplored.

Nonlinear magnetic dynamics have been widely investigated in other contexts, including magnon scattering and spin textures such as domain walls, vortices, and skyrmions²⁵⁻³². More broadly, frequency combs represent a hallmark of nonlinear coherent dynamics across the physical sciences. Building on the seminal work of Hall and Hänsch in optical frequency-comb metrology³³⁻³⁶, they provide a universal route to generating discrete high-order spectral structures from fundamental modes³⁷⁻⁴¹. In particular, frequency comb formation arising from nonlinear mode coupling has been explored mainly in the gigahertz (GHz) regime²⁶⁻³², where interactions between spin-wave modes or dynamic spin textures produce discrete spectral features. More recently, nonlinear magnon coupling has been extended into the THz regime in antiferromagnetic systems^{42,43}, indicating that ultrafast spin dynamics can support nonlinear processes at much higher frequencies. For spintronic THz emitters, such nonlinear operations

would be particularly significant, as they could transform broadband transient emission into coherent, high-order spectral structures. Despite these developments, the realization of THz-frequency combs based on spintronic emitters in magnetic multilayers remains unexplored.

In this work, we demonstrate that nonlinear THz frequency combs can emerge from ultrafast spin dynamics in nano-engineered magnetic multilayers. We further identify the physical origin of this comb formation, showing that it is closely linked to the spatiotemporal overlap of interfacial spin-dynamic modes. In particular, nonlinear coupling between spin-charge conversion and exchange-driven dynamics⁴⁴ gives rise to coherent higher-order spectral components that follow a universal comb relation. These results establish a physically transparent and experimentally accessible route toward nonlinear THz generation and frequency-comb formation in spintronic systems.

Spatial overlapping of THz emissions in nano-thin magnetic multilayer

Since nonlinear coupling between different spin modes can give rise to frequency comb behavior, we hypothesize that at least two intrinsic ultrafast spin-dynamic modes may nonlinearly couple within nano-thin magnetic multilayers to generate a frequency comb. As schematically illustrated in Fig. 1a, an ultrafast optical pulse can excite ultrafast spin current⁸⁻¹⁴ and ultrafast exchange dynamics⁴⁴ near the interface between ferromagnet (Co) and heavy metal (Pt or W). One THz mode (f_1) may originate from one of these mechanisms, while another mode (f_2) can also be excited with different spin dynamical features. In Fig. 1a, the upper ΔM_x scale schematically denotes a localized magnetization change near the Co/(Pt or W) interface induced by ultrafast spin current, whereas the lower M_x scale represents the magnetization profile associated with ultrafast exchange dynamics. When these interfacial responses spatially and temporally overlap within the nano-thin multilayer geometry, strong nonlinear interaction can occur, leading to a spintronic THz frequency comb described as $f_{(m)}^{comb} = f_1 + m f_2$ ($m = 1, 2, 3, \dots$). THz emission from multiple ultrafast spin-dynamic channels is realized within a confined multilayer geometry. Here, nano-thin magnetic multilayers are engineered to enable the coexistence and spatial overlap of spin-current and exchange-driven dynamics, which act as distinct modes for frequency-comb generation.

In the present study, two kinds of magnetic multilayer samples are prepared. The first series is [Pt/Co/MgO]_n with $n = 1, 2$, and 3 , where a ferromagnetic 2-nm Co layer is sandwiched by metallic 1.8-nm Pt and insulating 1.8-nm MgO layers. The second series is [Pt/Co/W/MgO]_n

with $n = 1, 2$, and 3 , where a ferromagnetic 2-nm Co layer is sandwiched between metallic 1.8-nm Pt and 1.8-nm W layers, with a protective capping layer of insulating 1.8-nm MgO. The Pt and W layers are only 1.8 nm thick, which is within the range of nanometer-scale spin-diffusion lengths reported for Pt thin films (2.8–3.9 nm)⁴⁵ and W (~ 3.5 nm)⁴⁶. This thickness scale supports interfacial spin-current and spin-charge-conversion processes while preserving nanoscale spatial overlap with the ultrafast exchange-driven dynamics in the adjacent Co layer. THz emission is generated from ultrafast spin dynamics in the Co layer as well as from interfacial spin-current and spin-charge conversion processes at the Pt/Co and Co/W interfaces. Both sample series exhibit in-plane magnetic anisotropy with similar coercivities, as evidenced by their magnetic hysteresis loops. (Supplementary Fig. S1). The sample structure is illustrated in the inset of Fig. 1b, while the optical-pump/THz-emission stroboscopy setup scheme is shown in the figure. Since the entire structures consist of nano-thin multilayers, the spatial overlap of ultrafast interfacial spin-dynamic responses is expected to be substantial, thereby enabling nonlinear interactions and the possible formation of frequency combs within the multilayers.

Frequency comb formation in [Pt/Co/MgO]_n

Figure 2 summarizes THz emission in [Pt/Co/MgO]_n multilayers with variation of pump fluence (F_P) as well as multilayer repeat number (n). Electric field profiles of THz emission waves are plotted in Fig. 2a and 2b for F_P of 0.8 and 1.6 mJ cm⁻², respectively. The emitted THz amplitude clearly increases with increasing pump fluence, as shown in the figure. The peak-to-peak THz amplitude for $n=3$ increases by factors of ~ 8 and ~ 30 compared to $n = 1$ and 2 , respectively. These behaviors become clearer in the Fourier spectra obtained via Fast Fourier Transform (FFT), as shown in Fig. 2c and 2d. Inset figures show the FFT spectra for $n = 1, 2$, and 3 , normalized for comparison in the range up to 3 THz. The peaks for $n = 1$ and 2 (~ 0.4 THz) shift to a higher frequency around ~ 0.8 THz for $n = 3$ (highlighted in the figure). The shift of the frequency peak to a higher value implies that the THz emission process changes as the number of interfaces increases from $n=1$ to $n=3$. Previous studies have shown that in ferromagnet(FM)/nonmagnetic(NM)-metal heterostructures, an excessively thin nonmagnetic layer may limit its effectiveness as a spin reservoir, thereby reducing the amount of injected spin current⁴⁷. In the [Pt/Co/MgO]_n series, increasing n effectively increases the number of spin-current-generating Pt/Co interfaces and the total Pt reservoir available for spin injection,

which may enhance the interfacial spin-current contribution. Also, the shift of the FFT peak toward ~ 0.8 THz at $n = 3$ suggests an enhanced contribution from ISHE.

FFT spectra on a log scale are plotted in Fig. 2e for the lower fluence ($F_P = 0.8 \text{ mJ cm}^{-2}$) over wide frequency ranges up to 15 THz. The spectra do not exhibit a dramatic difference overall, irrespective of n , whereas the subsequent change, as reflected in the observed peak shift below 3 THz, is evident. From 3 THz to 15 THz, the spectra appear almost flat, with no distinct structural features. However, it is interesting to note the difference in the FFT spectra shown in Fig. 2f for the higher-fluence case ($F_P = 1.6 \text{ mJ cm}^{-2}$). In the case of $n = 1$ and 2, the spectra exhibit no specific feature, as in the case of Fig. 2e. However, for $n = 3$, a clear periodic spectral structure emerges, as indicated by the green vertical bars. Another difference between Fig. 2e and 2f is that the amplitude of the spectra, only when the periodic structure emerges ($n = 3$ and $F_P = 1.6 \text{ mJ cm}^{-2}$), is significantly increased, hovering well above the flattened spectra in the 3–15 THz range, as observed in cases of $n = 1$ and 2. Specifically, the spectra without comb formation remain mostly at approximately -75 to -100 dB in the 3–15 THz range, whereas the comb-forming $n = 3$ spectrum exhibits periodic peaks around -30 to -60 dB, corresponding to more than one to two orders of magnitude enhancement in FFT amplitude. The observed periodic structure provides a clue to the expected frequency comb phenomenon, which is observed only at high F_P (1.6 mJ cm^{-2}) and with more interfaces ($n = 3$).

Frequency comb formation in $[\text{Pt/Co/W/MgO}]_n$

We have carried out the same experiment for the sample with an additional W layer inserted in the multilayer, $[\text{Pt/Co/W/MgO}]_n$. The increase of THz emission peak amplitude is similarly observed with increasing pump fluence from 0.8 to 1.6 mJ cm^{-2} , as shown in Fig. 3a and 3b. However, the main peaks are observed around ~ 0.8 THz for all cases, unlike those in $[\text{Pt/Co/MgO}]_n$, as shown by the FFT spectra in Fig. 3c and 3d. The inset figure clearly shows that the normalized peaks for multilayers with different n all coincide below 3 THz. This indicates that the THz emission is dominantly governed by the same generation mechanism. Here, it should be noted that the addition of the W layer enhanced the THz emission, supporting an ISHE-dominated contribution^{13,14}. In addition, a reduction in the THz amplitude from $n = 1$ to $n = 2$ is observed with the insertion of the W layer. A similar trend has been observed in photonic-crystal-type spintronic THz emitters, including $[\text{SiO}_2/\text{Pt/Fe/W}]_n$. It originates from

the competition between laser absorption enhancement and THz attenuation as the number of layers increases⁴⁸.

For the lower F_P (0.8 mJ cm^{-2}), the FFT spectra on a log scale do not exhibit a periodic structure, as seen in Fig. 3e. However, for the higher F_P (1.6 mJ cm^{-2}), periodic spectral structures are observed up to 15 THz for all values of n . In contrast to the $[\text{Pt/Co/MgO}]_n$ series, where the periodic feature appears only for $n = 3$ at $F_P = 1.6 \text{ mJ cm}^{-2}$, the W-inserted multilayers exhibit periodic spectral modulation for all n at high fluence (Fig. 3f). These results indicate that both higher pump fluence and W-layer insertion promote the formation of periodic spectral modulation in the emitted THz spectra.

The addition of the W layer is considered to enhance the spin-charge conversion efficiency, particularly via the inverse spin Hall effect, since the W has a negative spin Hall angle, whereas the Pt has a positive spin Hall angle^{13,14}. The spin current generated by the photoinduced ultrafast spin dynamics is pumped into the W as well as the Pt layer in the Pt/Co/W/MgO multilayer structure, where the opposite direction of spin currents from Co to Pt and W eventually induces the same direction of charge current due to the negative spin Hall angle of W, thereby inducing increased charge current. This behavior leads to stronger THz emission with increased amplitudes in the multilayer with W inserted (Fig. 2a,b vs. Fig. 3a,b). The resulting enhancement is consistent with ISHE-dominated interfacial spin-charge conversion, because without constructive spin-to-charge conversion from W, the additional metallic layer would be expected to increase attenuation rather than enhance the THz emission signal. Periodic frequency modulation emerges at large interface numbers and high pump fluence, marking the activation of nonlinear coupling between spin modes. The resulting regular spectral spacing signifies the formation of a frequency comb.

Analysis of frequency comb

To quantitatively confirm the frequency comb nature of the observed spectral peaks, the peak positions of the periodic spectral modulation at a pump fluence of 1.6 mJ cm^{-2} were extracted and plotted as a function of the integer comb index m (Fig. 4a). Remarkably, the peak frequencies obey a universal linear relation, as $f_{\text{comb}} = f_1 + mf_2$, where $f_1 \approx 0.95 \text{ THz}$ and $f_2 \approx 2.26 \text{ THz}$ are determined from the linear fitting. Note that the linear combination of the generated THz peaks is universal, regardless of the sample repetition number n or the presence of the W layer, once higher-order THz peaks are generated. This behavior indicates that the

comb spacing is governed primarily by intrinsic interfacial spin-dynamic modes rather than by structural repetition effects of the multilayer stack. The proportionality between f and m confirms that the regularly spaced peaks originate from coherent interaction involved with ultrafast photoinduced spin dynamics.

To analyze the origin of f_1 and f_2 modes, the THz emission signal of the four samples exhibiting high-order THz frequency peaks was carefully examined. To extract the fundamental modes of the signal, we applied the following fitting equation (Eq. (1)), composed of a Gaussian function multiplied by a cosine function.

$$E_y^{\text{mode}}[t] = G_{\text{mode}}[t] \cos[2\pi f_{\text{mode}}t - \varphi_b], \quad (1)$$

where $G_{\text{mode}}[t]$ represents the temporal envelope of Gaussian form and f_{mode} represents each fundamental mode. φ_b is a phase constant. From the fit, three distinct modes are found at approximately 0.4, 0.8, and 2.0 THz. The fitting parameters and their uncertainties are summarized in Supplementary Table S1. Three distinct modes are separated and then inverse Fourier transformed to visualize their time-domain components, as plotted in Fig. 4b for the [Pt/Co/W/MgO]₁ sample. FFT results for the separated modes are plotted in Fig. 4c, where it is observed that the mode of 0.8 THz is most dominant, followed by the mode of 0.4 THz and, lastly, by the mode of 2.0 THz. The same analysis was further extended to all four samples exhibiting frequency comb formation, and the resulting decomposed waveforms are summarized in Fig. 5a. The separated components near ~0.4, ~0.8, and ~2.0 THz are compared across the four samples in Fig. 5b-d, respectively, while the corresponding results at 0.8 mJ cm⁻² are provided in Fig. S2. It is observed that, in all cases of frequency combs, the ~0.8 THz mode is the most dominant one, whereas the ~2.0 THz mode remains weaker but is consistently resolved in every sample. Although the ~0.4 THz mode is also clearly present, the comb-scaling relation in Fig. 4a indicates that the dominant comb-relevant mode pair is associated primarily with the ~0.8 and ~2.0 THz components. We note that the fitted comb parameters $f_1 \approx 0.95$ THz and $f_2 \approx 2.26$ THz in Fig. 4a are slightly shifted from the nominal ~0.8 and ~2.0 THz mode centers obtained from the time-domain decomposition. This difference may arise from the finite temporal duration and spectral bandwidth of the ultrafast THz wave packets, since the comb relation is extracted from broadband pulsed spectra rather than isolated continuous-wave excitations.

The mode of 0.4 THz is considered to originate primarily from photoinduced magnetization dynamics⁵⁻⁷ associated with ultrafast demagnetization of the Co layer, since it is more clearly observable for the sample [Pt/Co/MgO]_n in case of n = 1 and 2, where the number of interfaces is limited and interfacial contributions are relatively weak. This mode becomes relatively less significant when the interfacial spin-charge conversion contribution is enhanced by inserting the W layer in the [Pt/Co/W/MgO]_n samples, as shown in Fig. 3. On the other hand, the ~0.8 THz mode is considered to originate from interfacial spin-charge conversion at the Co/Pt or Co/W interfaces. THz emission near 2.0 THz has also been observed in other photoinduced THz emission experiments and may be associated with exchange interaction breaking and recovery processes⁴⁴. We therefore associate the higher-frequency mode with strongly localized interfacial spin dynamics related to ultrafast exchange breaking and recovery near the Co/(Pt or W) interface. The dominant 0.8 THz emission component is attributed primarily to interfacial spin-charge conversion, most likely via the inverse spin Hall effect, as the signal is enhanced by inserting a W layer with an opposite spin Hall angle. This interfacial spin-charge conversion process is closely interrelated with both photoinduced magnetization dynamics and exchange interaction dynamics in the multilayer structure. It should be noted, however, that both temporal and spatial overlap are required for a frequency comb phenomenon to emerge.

Temporal overlap governing THz frequency comb formation

We have carried out further analysis based on Hilbert transform to isolate and analyze comb-building frequency components from the measured THz waveforms, as seen in Fig. 5. $E_y^{\text{mode}}[t]$ mode determined by Eq. (1) is processed by Hilbert transform⁴⁹ as in Eq. (2) to form a complex $\tilde{E}_y^{\text{mode}}[t]$.

$$\tilde{E}_y^{\text{mode}}[t] = E_y^{\text{mode}}[t] + i \mathcal{H} [E_y^{\text{mode}}[t]], \quad (2)$$

where $\mathcal{H}$ is the Hilbert transform and the mode is for 0.4, 0.8, and 2.0 THz. Then, the wave envelope is determined as Eq. (3):

$$|\tilde{E}_y^{\text{mode}}[t]| = \sqrt{\tilde{E}_y^{\text{mode}}[t] \tilde{E}_y^{\text{mode}*}[t]}, \quad (3)$$

where $\tilde{E}_y^{\text{mode}*}[t]$ denotes the complex conjugate. The measured field is $\text{Re} [\tilde{E}_y^{\text{mode}}[t]]$.

The estimated wave envelopes are plotted for the case of four samples in Fig. 6a for the case of $F_P = 1.6 \text{ mJ cm}^{-2}$. The envelope is again decomposed into $\sim 0.4 \text{ THz}$ (black), $\sim 0.8 \text{ THz}$ (red), and $\sim 2.0 \text{ THz}$ (blue), with the overlap region colored green. Among the possible pairwise overlap channels, the overlap between the $\sim 0.8 \text{ THz}$ and $\sim 2.0 \text{ THz}$ envelopes is highlighted because Fig. 4a identifies these two modes as the principal comb-relevant pair. The temporal overlap between the $\sim 0.8 \text{ THz}$ and $\sim 2.0 \text{ THz}$ envelopes defines the interaction window. Within this window, nonlinear coupling leading to frequency-comb formation can be phenomenologically described as⁵⁰

$$\tilde{E}_y^{\text{comb}}[t] \approx \chi^{(2)} \tilde{E}_y^{0.8\text{THz}}[t] \tilde{E}_y^{2.0\text{THz}}[t], \quad (4),$$

and the overlap envelope, as colored green in the figure, enabling the frequency comb is estimated by

$$|\tilde{E}_y^{\text{comb}}[t]| \approx |\chi^{(2)}| |\tilde{E}_y^{0.8\text{THz}}[t] \tilde{E}_y^{2.0\text{THz}}[t]|. \quad (5)$$

Here, $\chi^{(2)}$ is introduced as an effective phenomenological coefficient representing the nonlinear interaction strength between the two modes. For the present comparison, $\chi^{(2)}$ is set to unity, so that the overlap-derived waveform is used only to compare the relative temporal profile rather than to quantify the absolute nonlinear coupling strength. Multiple overlap channels may in principle coexist and contribute to the measured comb waveform; however, the present analysis indicates that the $\sim 0.8 \text{ THz} \otimes \sim 2.0 \text{ THz}$ channel provides the dominant contribution to the observed frequency comb formation, likely because this interaction channel more effectively satisfies the spatiotemporal overlap condition at the ferromagnet/heavy-metal (FM/HM, here Co/Pt or Co/W) interface. Because the measured comb waveform may contain secondary contributions from additional overlap channels, the effective amplitude of the overlap-derived packet should not be interpreted as a direct measure of a single nonlinear coefficient. Nevertheless, the present comparison indicates that the $\sim 0.8 \text{ THz} \otimes \sim 2.0 \text{ THz}$ channel captures the dominant temporal structure of the observed comb waveform.

It is worth noting that the wave packet for the $\sim 0.8 \text{ THz}$ mode appears earlier than that for

the ~ 2.0 THz mode by several hundred femtoseconds, as seen in Fig. 6a. This indicates that the physical process corresponding to the 0.8 THz peak, which is associated with the inverse spin Hall effect, occurs earlier in time, followed by the physical process corresponding to the 2.0 THz peak, which is associated with exchange breaking and recovery. The wave packet for the 0.4 THz peak is also shown in Fig. 6a, where it exhibits a broader temporal envelope than the higher-frequency components. Although it is widely distributed in time, the comb behavior identified in Fig. 4a and the comparison of the separated modes in Fig. 5 suggest that this component does not play a dominant role in the main comb-forming channel. This low-frequency component may be associated with ultrafast demagnetization in the Co layer or with spin-current dynamics originating in the bulk rather than at the interface. As such, it may not effectively satisfy the spatial overlap condition at the FM/HM interface, which is expected to be essential for nonlinear mode coupling and the formation of a densely packed frequency comb.

The high-order frequency profiles can be extracted from the residual signal (Exp. - Fit.), as shown in Fig. 6b. Since only the low frequency modes of 0.4, 0.8, and 2.0 THz are removed through the fitting procedure, the remaining signal is dominated by the high-frequency comb components. The high-frequency comb signal is shown together with an overlapping envelope, $\tilde{E}_y^{\text{comb}}[t]$, where the high-frequency comb components are found to be consistently matched with a temporally overlapped region. This comparison supports the view that the dominant comb signal emerges within the same temporal window where the ~ 0.8 THz and ~ 2.0 THz wave packets coexist.

For the case without frequency-comb formation, a similar analysis was carried out, as seen in Fig. 6c-d. As clearly seen in the figure, the temporal overlapping region is much smaller than in Fig. 6a, implying that the temporal overlapping window is only fractional, discouraging possible frequency comb formation. Another interesting feature in this case is that only the 0.8 THz mode is dominant, with the other modes almost negligible and thus insufficient to combine into a frequency comb. The high-order oscillation, determined by subtracting $E_y^{\text{mode}}[t]$ from the measured E_y , is also found to be negligible as in Fig. 6d. This result is consistent with the view that, under weak excitation, the temporal interaction window is too limited to support an efficient nonlinear coupling channel.

High-order components of THz frequency comb

To examine the structure of the THz frequency comb, the high-order components extracted from the measured $E_y[t]$ at a pump fluence of 1.6 mJ cm^{-2} (“Exp. – Fit.” in Fig. 6b) were further analyzed. Owing to the experimental time resolution of 300 fs, frequency components up to $\sim 16 \text{ THz}$ are accessible, and band-pass filtering was applied to isolate the comb components up to the 6th order ($m = 1 \sim 6$ in $f_{(m)}^{comb} = f_1 + mf_2$). All extracted components up to the 6th order were analyzed using a band-pass filter with a full width at half maximum (FWHM) of 1.1 THz , determined from the experimentally measured spectral bandwidth. The 1.1 THz FWHM is less than half the comb spacing ($\sim 2.26 \text{ THz}$), ensuring adequate spectral selectivity between adjacent orders. The corresponding results for the low-fluence condition (0.8 mJ cm^{-2}) are presented in Fig. S5 of the Supplementary Information.

In Fig. 7a, the experimental signal and the reconstructed waveform based on the extracted comb components are plotted together, showing good agreement with the observed temporal modulation. Figures 7b and 7c highlight the first- and second-order ($m = 1$ and 2) components of the frequency comb. The lowest-order component ($m = 1$) exhibits the strongest amplitude and a well-defined wave packet, indicating a dominant nonlinear contribution, while the second-order component ($m = 2$) also shows a pronounced amplitude, confirming strong nonlinear coupling between the underlying modes. In contrast, the signal becomes noticeably weaker for higher orders ($m \geq 3$), where only the comb-like spacing remains discernible. This indicates that the frequency comb behavior persists in the higher-order regime, although with substantially reduced contribution from higher-order components.

Nevertheless, components up to $m = 6$ can still be identified within the accessible spectral range, suggesting that the comb structure extends beyond the dominant low-order regime. All extracted components exhibit wave-packet envelopes $\sim 2 \text{ ps}$, highlighting that temporal overlap, in addition to spatial overlap, plays an important role in the formation of the THz frequency comb.

Conclusions

In summary, we have experimentally demonstrated the emergence of a THz frequency comb in spintronic magnetic multilayers. By engineering $[\text{Pt/Co/W/MgO}]_n$ heterostructures to maximize interfacial spin-charge conversion, we have successfully moved beyond the linear regime of THz emission into a regime of coherent nonlinear dynamics. Our findings reveal that the frequency comb is not merely a high-power artifact, but a precisely structured phenomenon

arising from the nonlinear coupling of two fundamental spin modes, specifically an ISHE-dominated spin–charge conversion mode (~ 0.8 THz) and exchange-interaction dynamics (~ 2.0 THz). The universal linear scaling of the comb-order peak positions, $f_{(m)}^{comb} = f_1 + mf_2$, confirms that these periodic spectral structures are fundamentally linked to the sub-picosecond temporal overlap and nanoscale spatial confinement of the excitation packets. The addition of a W layer, whose spin Hall angle is opposite to that of Pt, significantly enhances the interfacial spatiotemporal overlap between the comb-relevant modes, thereby facilitating the generation of higher-order THz comb components up to 15 THz. This work connects nonlinear frequency-comb physics with spintronic THz emission, establishing magnetic multilayers as a versatile platform for coherent high-order THz spectral structuring. Beyond fundamental interest, these results provide a new spectroscopic ruler for measuring and manipulating ultrafast electron, lattice, and spin interactions on their intrinsic non-equilibrium timescales. Our approach provides a foundation for high-precision spintronic metrology and, with further engineering, may enable compact THz frequency-comb sources for spectroscopy and communication.

Method

THz emission experiment:

The THz emission measurements were conducted at the Pohang Accelerator Laboratory (PAL) in South Korea. A femtosecond laser pulse was generated by an amplified Ti:sapphire laser with a central wavelength of 800 nm, a bandwidth of 20 nm, a pulse duration of 120 fs, and a repetition rate of 1 kHz. The pump pulse was typically incident on the sample surface at a fluence of 0.8 or 1.6 mJ cm⁻², with an approximate spot diameter of 5 mm at the e^{-2} intensity level. In the optical pump-probe experiment, both the pump and probe pulses were p-polarized. A 1-mm-thick ZnTe crystal was used to detect the THz signal via electro-optic sampling. The time-domain THz emission experiment was performed using a femtosecond-laser pump in an in-plane external magnetic field of 30 mT. The schematic diagram of the experimental setup is shown in Fig. 1b. All time-domain THz waveforms were obtained by averaging the results of three or more repeated scans under identical experimental conditions. The individual scans showed nearly identical temporal profiles, confirming the high reproducibility of the measured THz emission signals.

Sample Fabrication:

The multilayer samples [Pt(1.8)/Co(2)/MgO(1.8)]_n and [Pt(1.8)/Co(2)/W(1.8)/MgO(1.8)]_n were grown on 0.5-mm-thick, double-polished, (0001)-oriented sapphire substrates by magnetron sputtering. All films were deposited at room temperature after achieving a base pressure of $\sim 10^{-8}$ Torr. The Pt layer was deposited at a working pressure of 0.593 mTorr with a thickness of 1.8 nm. The Co layer was deposited at a working pressure of 0.578 mTorr with a thickness of 2 nm. The W layer was deposited at a working pressure of 0.593 mTorr with a thickness of 1.8 nm. The MgO layer, used as a capping layer to prevent oxidation, was deposited at a working pressure of 4.125 mTorr with a thickness of 1.8 nm.

Data availability

The data supporting the findings of this study, including the numerical data underlying the figures, are available from the corresponding authors upon reasonable request.

Acknowledgments

This research was supported by the National Natural Science Foundation of China (Grant No. 12364020) and the Natural Science Foundation of Jilin Province [Grant No. 20240101295JC]. This research was also supported by the National Research Foundation of Korea (NRF) (grant Nos. RS-2023-00250178 and RS-2025-25433745). This work was supported by IITP (Institute of Information & Communications Technology Planning & Evaluation)-ITRC (Information Technology Research Center) grant funded by the Korea government (Ministry of Science and ICT) (IITP-2025-RS-2024-00437284). This research was also supported by the Regional Innovation System & Education (RISE) program through the Chungbuk Regional Innovation System & Education Center, funded by the Ministry of Education (MOE) and the Chungcheongbuk-do, Republic of Korea (2025-RISE-11-014-05). This research was also supported by a Global-Learning & Academic research institution for Master's-Ph.D. students, and Postdocs (LAMP) Program of the National Research Foundation of Korea (NRF) grant funded by the Ministry of Education (Grant No. RS-2024-00445180).

Author Contribution

J.-H.S. and Y.Z. contributed equally to this work. They conceived the study, designed the experimental framework, analyzed the data, interpreted the results, and prepared the manuscript. Q.M., M.J., J.H., P.S., and D.P. carried out the THz emission measurements and contributed to data acquisition. H.J.S. and J.H.P. provided the THz experimental setup and technical support at PAL. X.Y. contributed to discussions and interpretation of the experimental results. S.X., C.W., and X.H. fabricated multilayer samples and performed their structural and magnetic characterizations. K.W.K., C.W., H.-G.P., and D.-H.K. provided scientific guidance on the experimental strategy and interpretation of the results and supervised the overall project. All authors discussed the results and commented on the manuscript.

Additional Information

Supplementary information: The online version contains supplementary material available at <https://doi.org/>.

Competing financial interests: The authors declare no competing financial interests.

References

1. Sirtori, C. Bridge for the terahertz gap. *Nature* **417**, 132–133 (2002).
2. Ferguson, B. & Zhang, X.-C. Materials for terahertz science and technology. *Nat. Mater.* **1**, 26–33 (2002).
3. Tonouchi, M. Cutting-edge terahertz technology. *Nat. Photonics* **1**, 97–105 (2007).
4. Hoffmann, M. C. & Fülöp, J. A. Intense ultrashort terahertz pulses: generation and applications. *J. Phys. D: Appl. Phys.* **44**, 083001 (2011).
5. Beaurepaire, E., Turner, G. M., Harrel, S. M., Beard, M. C., Bigot, J.-Y. & Schmuttenmaer, C. A. Coherent terahertz emission from ferromagnetic films excited by femtosecond laser pulses. *Appl. Phys. Lett.* **84**, 3465 (2004).
6. Huang, L., Kim, J.-W., Lee, S.-H., Kim, S.-D., Tien, V. M., Shinde, K. P., Shim, J.-H., Shin, Y., Shin, H. J., Kim, S., Park, J., Park, S.-Y., Choi, Y. S., Hong, J.-I. & Kim, D.-H. Direct observation of terahertz emission from ultrafast spin dynamics in thick ferromagnetic films. *Appl. Phys. Lett.* **115**, 142404 (2019).
7. Huang, L., Lee, S.-H., Kim, S.-D., Shim, J.-H., Shin, H. J., Kim, S., Park, J., Park, S.-Y., Choi, Y. S., Kim, H.-J., Hong, J.-I., Kim, D.-E. & Kim, D.-H. Universal field-tunable terahertz emission by ultrafast photoinduced demagnetization in Fe, Ni and Co ferromagnetic films. *Sci. Rep.* **10**, 15843 (2020).
8. Kampfrath, T., Battiato, M., Maldonado, P., Eilers, G., Nötzold, J., Mährlein, S., Zbarsky, V., Freimuth, F., Mokrousov, Y., Blügel, S., Wolf, M., Radu, I., Oppeneer, P. M. & Münzenberg, M. Terahertz spin current pulses controlled by magnetic heterostructures. *Nat. Nanotechnol.* **8**, 256 (2013).
9. Seifert, T., Jaiswal, S., Martens, U., Hannegan, J., Braun, L., Maldonado, P., Freimuth, F., Kronenberg, A., Henrizi, J., Radu, I., Beaurepaire, E., Mokrousov, Y., Oppeneer, P. M., Jourdan, M., Jakob, G., Turchinovich, D., Hayden, L. M., Wolf, M., Münzenberg, M., Kläui, M. & Kampfrath, T. Efficient metallic spintronic emitters of ultrabroadband terahertz radiation. *Nat. Photonics* **10**, 483 (2016).
10. Seifert, T., Martens, U., Günther, S., Schoen, M. A. W., Radu, F., Chen, X. Z., Lucas, I., Ramos, R., Aguirre, M. H., Algarabel, P. A., Anadón, A., Körner, H. S., Walowski,

- J., Back, C., Ibarra, M. R., Morellón, L., Saitoh, E., Wolf, M., Song, C., Uchida, K., Münzenberg, M., Radu, I. & Kampfrath, T. Terahertz spin currents and inverse spin Hall effect in thin-film heterostructures containing complex magnetic compounds. *Spin* **7**, 1740010 (2017).
11. Cheng, L., Li, Z., Zhao, D. & Chia, E. E. M. Studying spin–charge conversion using terahertz pulses. *APL Mater.* **9**, 070902 (2021).
 12. Ilyakov, I., Brataas, A., de Oliveira, T. V. A. G., Ponomaryov, A., Deinet, J.-C., Hellwig, O., Faßbender, J., Lindner, J., Salikhov, R. & Kovalev, S. Efficient ultrafast field-driven spin current generation for spintronic terahertz frequency conversion. *Nat. Commun.* **14**, 7010 (2023).
 13. Wu, X., Wang, H., Liu, H., Wang, Y., Chen, X., Chen, P., Li, P., Han, X., Miao, J., Yu, H., Wan, C., Zhao, J. & Chen, S. Antiferromagnetic–ferromagnetic heterostructure-based field-free terahertz emitters. *Adv. Mater.* **34**, 2204373 (2022).
 14. Zhao, Y., Gayen, A., Huang, L., You, X., Le Thi, N., Mustaghfiroh, Q., Rahmani, F., Gaikwad, P. V., Yen, P. D. H., Shim, J.-H., Kim, D. E., Shin, H. J., Park, J., Wan, C., Xiang, O., Piao, H.-G., Kim, K.-H., Bang, J., Lee, H. S., Kim, K. W. & Kim, D.-H. Quantifying spin-charge conversion mechanisms for THz emission in magnetic multilayers. *Adv. Optical Mater.* **12**, 2302571 (2024).
 15. Zhou, C., Liu, Y. P., Wang, Z., Ma, S. J., Jia, M. W., Wu, R. Q., Zhou, L., Zhang, W., Liu, M. K., Wu, Y. Z. & Qi, J. Broadband terahertz generation via the interface inverse Rashba–Edelstein effect. *Phys. Rev. Lett.* **121**, 086801 (2018).
 16. Jungfleisch, M. B., Zhang, Q., Zhang, W., Pearson, J. E., Schaller, R. D., Wen, H. & Hoffmann, A. Control of terahertz emission by ultrafast spin–charge current conversion at Rashba interfaces. *Phys. Rev. Lett.* **120**, 207207 (2018).
 17. Liu, X., Chanana, A., Huynh, U., Xue, F., Haney, P., Blair, S., Jiang, X. & Vardeny, Z. V. Circular photogalvanic spectroscopy of Rashba splitting in 2D hybrid organic–inorganic perovskite multiple quantum wells. *Nat. Commun.* **11**, 323 (2020).
 18. Huisman, T. J., Mikhaylovskiy, R. V., Costa, J. D., Freimuth, F., Paz, E., Ventura, J., Freitas, P. P., Blügel, S., Mokrousov, Y., Rasing, Th. & Kimel, A. V. Femtosecond control of electric currents in metallic ferromagnetic heterostructures. *Nat.*

Nanotechnol. **11**, 455–458 (2016).

19. Huang, L., Cao, Y., Qiu, H., Bai, H., Liao, L., Chen, C., Han, L., Pan, F., Jin, B. & Song, C. Terahertz oscillation driven by optical spin–orbit torque. *Nat. Commun.* **15**, 7227 (2024).
20. Zhang, Q., Luo, Z., Li, H., Yang, Y., Zhang, X. & Wu, Y. Terahertz emission from anomalous Hall effect in a single-layer ferromagnet. *Phys. Rev. Appl.* **12**, 054027 (2019).
21. Seifert, T. S., Martens, U., Radu, F., Ribow, M., Berritta, M., Nádvorník, L., Starke, R., Jungwirth, T., Wolf, M., Radu, I., Münzenberg, M., Oppeneer, P. M., Woltersdorf, G. & Kampfrath, T. Frequency-independent terahertz anomalous Hall effect in DyCo₅, Co₃₂Fe₆₈, and Gd₂₇Fe₇₃ thin films from DC to 40 THz. *Adv. Mater.* **33**, 2007398 (2021).
22. Cheng, H., Wang, Y., Liu, Z., Jia, X., Huang, Q. & Lu, Y. Terahertz spin-to-charge conversion in ferromagnetic Ni nanofilms. *Nanophotonics* **12**, 2145–2156 (2023).
23. Mandal, R., Momma, R., Ishibashi, K., Iihama, S., Suzuki, K. & Mizukami, S. Topologically influenced terahertz emission in Co₂MnGa with a large anomalous Hall effect. *NPG Asia Mater.* **16**, 30 (2024).
24. Battiato, M., Carva, K. & Oppeneer, P. M. Superdiffusive spin transport as a mechanism of ultrafast demagnetization. *Phys. Rev. Lett.* **105**, 027203 (2010).
25. Yao, X., Jin, Z., Wang, Z., Zeng, Z. & Yan, P. Terahertz magnon frequency comb. *Phys. Rev. B* **108**, 134427 (2023).
26. Zhou, Z.-W., Wang, X.-G., Nie, Y.-Z., Xia, Q.-L. & Guo, G.-H. Spin wave frequency comb generated through interaction between propagating spin wave and oscillating domain wall. *J. Magn. Magn. Mater.* **534**, 168046 (2021).
27. Yang, X., Li, H., Zhang, X.-F., Ma, X.-P., Shim, J.-H., Jin, Y. & Piao, H.-G. Directly tunable magnon frequency comb effect based on domain wall. *Chin. Phys. B* **34**, 107507 (2025).
28. Yang, X., Li, H., Zhang, X.-F., Ma, X.-P., Shim, J.-H., Wang, H.-F., Zhang, S. & Piao, H.-G. Rationally phase-locked magnon frequency combs from the pinned domain wall. *Appl. Phys. Lett.* **127**, 202404 (2025).

29. Schultheiss, K., Verba, R., Wehrmann, F., Wagner, K., Körber, L., Hula, T., Hache, T., Kákay, A., Awad, A. A., Tiberkevich, V., Slavin, A. N., Fassbender, J. & Schultheiss, H. Excitation of whispering gallery magnons in a magnetic vortex. *Phys. Rev. Lett.* **122**, 097202 (2019).
30. Wang, Z., Yuan, H. Y., Cao, Y. & Yan, P. Twisted magnon frequency comb and Penrose superradiance. *Phys. Rev. Lett.* **129**, 107203 (2022).
31. Wang, Z., Yuan, H. Y., Cao, Y., Li, Z.-X., Duine, R. A. & Yan, P. Magnonic frequency comb through nonlinear magnon–skyrmion scattering. *Phys. Rev. Lett.* **127**, 037202 (2021).
32. Liu, X., Jin, Z., Li, Z., Zeng, Z., Li, M., Yao, Y., Cao, Y., Zhang, Y. & Yan, P. Low-lying magnon frequency comb in skyrmion crystals. *Phys. Rev. B* **110**, 184413 (2024).
33. Jones, D. J., Diddams, S. A., Ranka, J. K., Stentz, A., Windeler, R. S., Hall, J. L. & Cundiff, S. T. Carrier-envelope phase control of femtosecond mode-locked lasers and direct optical frequency synthesis. *Science* **288**, 635–639 (2000).
34. Holzwarth, R., Udem, Th., Hänsch, T. W., Knight, J. C., Wadsworth, W. J. & Russell, P. St. J. Optical frequency synthesizer for precision spectroscopy. *Phys. Rev. Lett.* **85**, 2264–2267 (2000).
35. Hall, J. L. Nobel Lecture: Defining and measuring optical frequencies. *Rev. Mod. Phys.* **78**, 1279–1295 (2006).
36. Hänsch, T. W. Nobel Lecture: Passion for precision. *Rev. Mod. Phys.* **78**, 1297–1309 (2006).
37. Cundiff, S. T. & Ye, J. Colloquium: Femtosecond optical frequency combs. *Rev. Mod. Phys.* **75**, 325 (2003).
38. Kippenberg, T. J., Holzwarth, R. & Diddams, S. A. Microresonator-based optical frequency combs. *Science* **332**, 555–559 (2011).
39. Ludlow, A. D., Boyd, M. M., Ye, J., Peik, E. & Schmidt, P. O. Optical atomic clocks. *Rev. Mod. Phys.* **87**, 637 (2015).
40. Picqué, N. & Hänsch, T. W. Frequency comb spectroscopy. *Nat. Photonics* **13**, 146–157 (2019).

41. Fortier, T. & Baumann, E. 20 years of developments in optical frequency comb technology and applications. *Commun. Phys.* **2**, 153 (2019).
42. Zhang, Z., Gao, F. Y., Curtis, J. B., Liu, Z.-J., Chien, Y.-C., von Hoegen, A., Wong, M. T., Kurihara, T., Suemoto, T., Narang, P., Baldini, E. & Nelson, K. A. Terahertz field-induced nonlinear coupling of two magnon modes in an antiferromagnet. *Nat. Phys.* **20**, 801–806 (2024).
43. Huang, C., Luo, L., Mootz, M., Shang, J., Man, P., Su, L., Perakis, I. E., Yao, Y. X., Wu, A. & Wang, J. Extreme terahertz magnon multiplication induced by resonant magnetic pulse pairs. *Nat. Commun.* **15**, 3214 (2024).
44. Shim, J.-H., Zhao, Y., Mustaghfiroh, Q., Le Thi, N., Rahmani, F., Kim, K., Shin, H. J., Park, J., You, X., Wan, C., Jung, M.-S., Hong, J.-I., Piao, H.-G., Han, X. & Kim, D.-H. Terahertz emission modulation caused by ultrafast breaking and recovery of exchange bias. *Adv. Optical Mater.* **14**, e03272 (2026).
45. Nguyen, M.-H., Ralph, D. C. & Buhrman, R. A. Spin torque study of the spin Hall conductivity and spin diffusion length in platinum thin films with varying resistivity. *Phys. Rev. Lett.* **116**, 126601 (2016).
46. Hao, Q., Chen, W. & Xiao, G. Beta (β) tungsten thin films: Structure, electron transport, and giant spin Hall effect. *Appl. Phys. Lett.* **106**, 182403 (2015).
47. Chen, Z., Luo, J.-W. & Wang, L.-W. Light-induced ultrafast spin transport in multilayer metallic films originates from sp–d spin exchange coupling. *Sci. Adv.* **9**, eadi1618 (2023).
48. Feng, Z., Yu, R., Zhou, Y., Lu, H., Tan, W., Deng, H., Liu, Q., Zhai, Z., Zhu, L., Cai, J., Miao, B. & Ding, H. Highly efficient spintronic terahertz emitter enabled by metal–dielectric photonic crystal. *Adv. Opt. Mater.* **6**, 1800965 (2018).
49. Bracewell, R. N. *The Fourier Transform and Its Applications*, 3rd edn. (McGraw–Hill, New York, 2000).
50. Shen, Y. R. *The Principles of Nonlinear Optics*. (Wiley-Interscience, 1984).

Figures

Fig. 1 | Conceptual scheme and experimental setup. **a**, Schematic illustration of the proposed THz frequency comb formation in spintronic heterostructures. An ultrafast spin current response and ultrafast exchange dynamics coexist near the Co/(Pt or W) interface, and their spatiotemporal overlap gives rise to comb-forming nonlinear coupling that follows the relation $f_{(m)}^{comb} = f_1 + mf_2$. **b**, Experimental setup for femtosecond optical-pump/THz emission measurements on [Pt/Co/MgO]_n and [Pt/Co/W/MgO]_n multilayers.

Fig. 2 | Fluence-dependent THz emission from [Pt/Co/MgO]_n multilayers. **a,b**, Time-domain THz emission signals measured at pump fluences of 0.8 and 1.6 mJ cm⁻², respectively, for n = 1, 2, and 3. **c-d**, Corresponding FFT spectra on a linear scale. Insets show normalized FFT spectra below 3 THz, highlighting the evolution of low-frequency components; the shaded orange band marks the ~0.8 THz region, which becomes pronounced only for n = 3. **e-f**, FFT spectra on a log scale. While no distinct high-frequency features appear above 3 THz at low fluence, regularly spaced comb-like spectral lines emerge at 1.6 mJ cm⁻² for the n = 3 multilayer (green guides), indicating the onset of nonlinear THz frequency comb formation.

Fig. 3 | Fluence-dependent THz emission from [Pt/Co/W/MgO]_n multilayers. **a,b**, Time-domain THz emission signals measured at pump fluences of 0.8 and 1.6 mJ cm⁻², respectively, for n = 1, 2, and 3. **c-d**, Corresponding FFT spectra on a linear scale. Insets show normalized FFT spectra below 3 THz for direct comparison of the low-frequency components; the shaded orange band marks the ~0.8 THz region. **e-f**, FFT spectra on a log scale. While no distinct high-frequency features appear above 3 THz at low fluence, regularly spaced comb-like spectral lines emerge at 1.6 mJ cm⁻² for all [Pt/Co/W/MgO]_n multilayers (green guides).

Fig. 4 | Verification of THz frequency comb formation and mode decomposition. **a**, Peak positions extracted from the FFT spectra of [Pt/Co/MgO]₃ and [Pt/Co/W/MgO]_n multilayers at $F_p = 1.6$ mJ cm⁻². The observed peaks follow the linear relation $f_{(m)}^{comb} = f_1 + mf_2$, with $f_1 \approx 0.95$ THz and $f_2 \approx 2.26$ THz, confirming the discrete comb spacing. **b**, Representative mode decomposition of the THz emission waveform measured from the [Pt/Co/W/MgO]₁ sample at $F_p = 1.6$ mJ cm⁻², obtained using Eq. (1). The experimental waveform (open black symbols) is

reproduced by a superposition of three characteristic components centered near: ~ 0.4 THz (black), ~ 0.8 THz (red), and ~ 2.0 THz (blue). The green dashed curve denotes the total fitted waveform. **c**, FFT spectra corresponding to the decomposed time-domain components shown in **b**, together with the experimental spectrum and the total fit.

Fig. 5 | Representative time-domain decomposition of comb-relevant THz modes at high fluence. High-fluence results (1.6 mJ cm^{-2}) for $[\text{Pt/Co/MgO}]_3$, $[\text{Pt/Co/W/MgO}]_1$, $[\text{Pt/Co/W/MgO}]_2$, and $[\text{Pt/Co/W/MgO}]_3$. **a**, Experimental THz waveforms (open squares), total fitted signals (green dashed lines), decomposed mode components (~ 0.4 THz, black; ~ 0.8 THz, red; ~ 2.0 THz, blue), and the residual component obtained from $\text{Exp.} - \text{Fit.}$ (magenta). **b–d**, Individual time-domain components centered near ~ 0.4 THz, ~ 0.8 THz, and ~ 2.0 THz, respectively. Solid lines show the oscillatory waveforms, and dashed lines denote their corresponding envelopes.

Fig. 6 | Temporal overlap of interfacial THz modes and emergence of the comb-related wave packet. **a,b**, High-fluence results (1.6 mJ cm^{-2}) for $[\text{Pt/Co/MgO}]_3$ and $[\text{Pt/Co/W/MgO}]_n$ multilayers. **a**, Temporal envelopes of the ~ 0.4 THz (black), ~ 0.8 THz (red), and ~ 2.0 THz (blue) components. The shaded green region indicates the overlap envelope $|\tilde{E}_y^{\text{comb}}[t]|$ obtained from the combined temporal overlap of the higher-frequency modes. **b**, Corresponding residual signals ($\text{Exp.} - \text{Fit.}$) together with the extracted comb-related wave packet (green shaded region), showing strong temporal correlation with the overlap envelope. **c,d**, Same analysis as in **a,b**, respectively, performed at a lower pump fluence of 0.8 mJ cm^{-2} , where the overlap envelope and the corresponding comb-related signal are strongly suppressed.

Fig. 7 | Extraction of individual comb orders from the measured THz waveforms.

a, Experimental residual signals ($\text{Exp.} - \text{Fit.}$, black squares) at high pump fluence (1.6 mJ cm^{-2}) for $[\text{Pt/Co/MgO}]_3$, and $[\text{Pt/Co/W/MgO}]_n$ multilayers, together with reconstructed traces (orange) obtained by summing the extracted comb-order components. **b–g**, Band-pass-filtered comb-order components corresponding to $m = 1\sim 6$, with center frequencies $f_{(m)}^{\text{comb}} = 3.21, 5.47, 7.73, 9.99, 12.25, \text{ and } 14.51 \text{ THz}$, respectively. The higher-order components ($m \geq 3$) are vertically scaled for clarity.

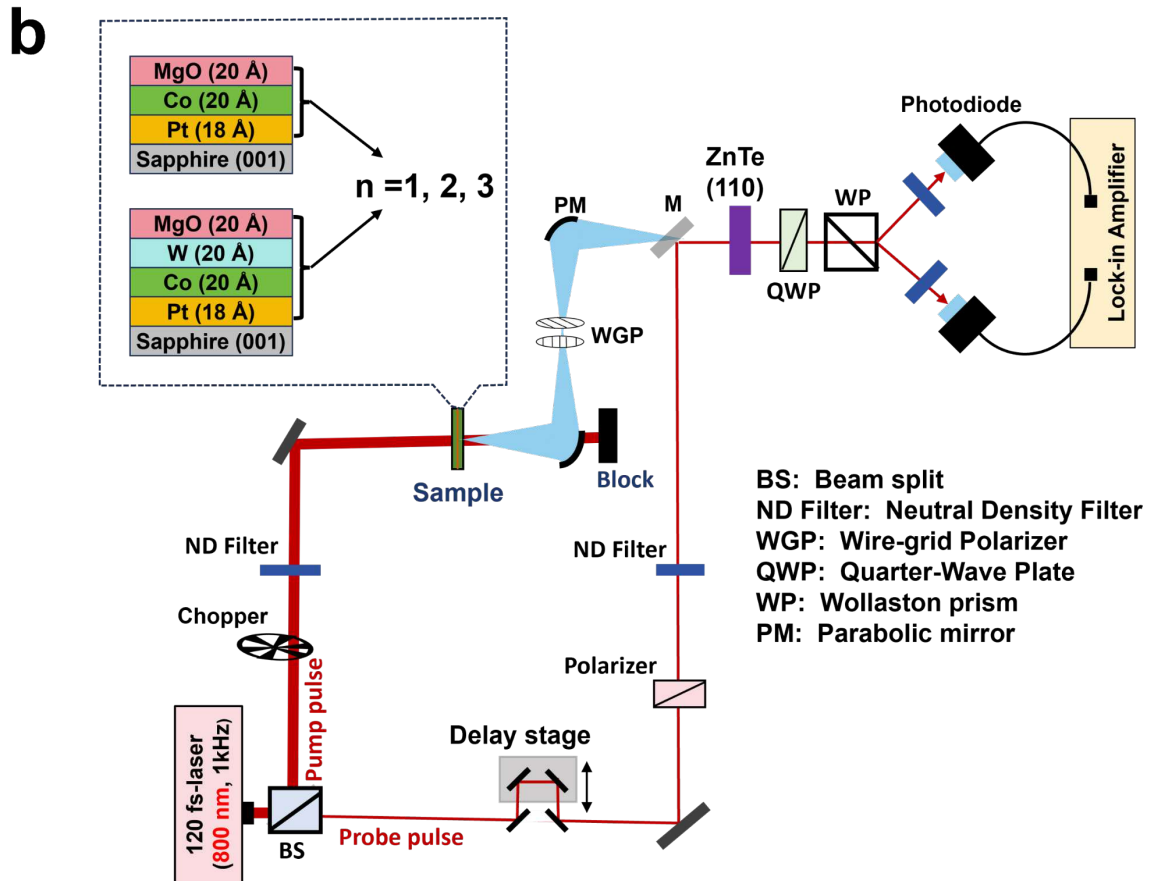
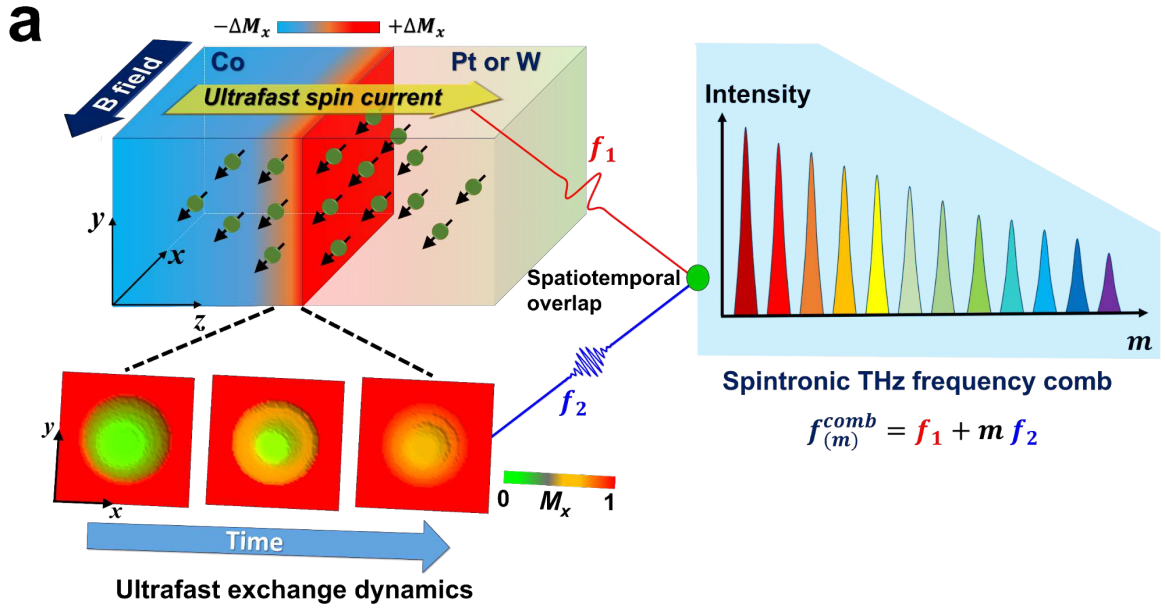


Fig. 1

[Pt/Co/MgO]_n

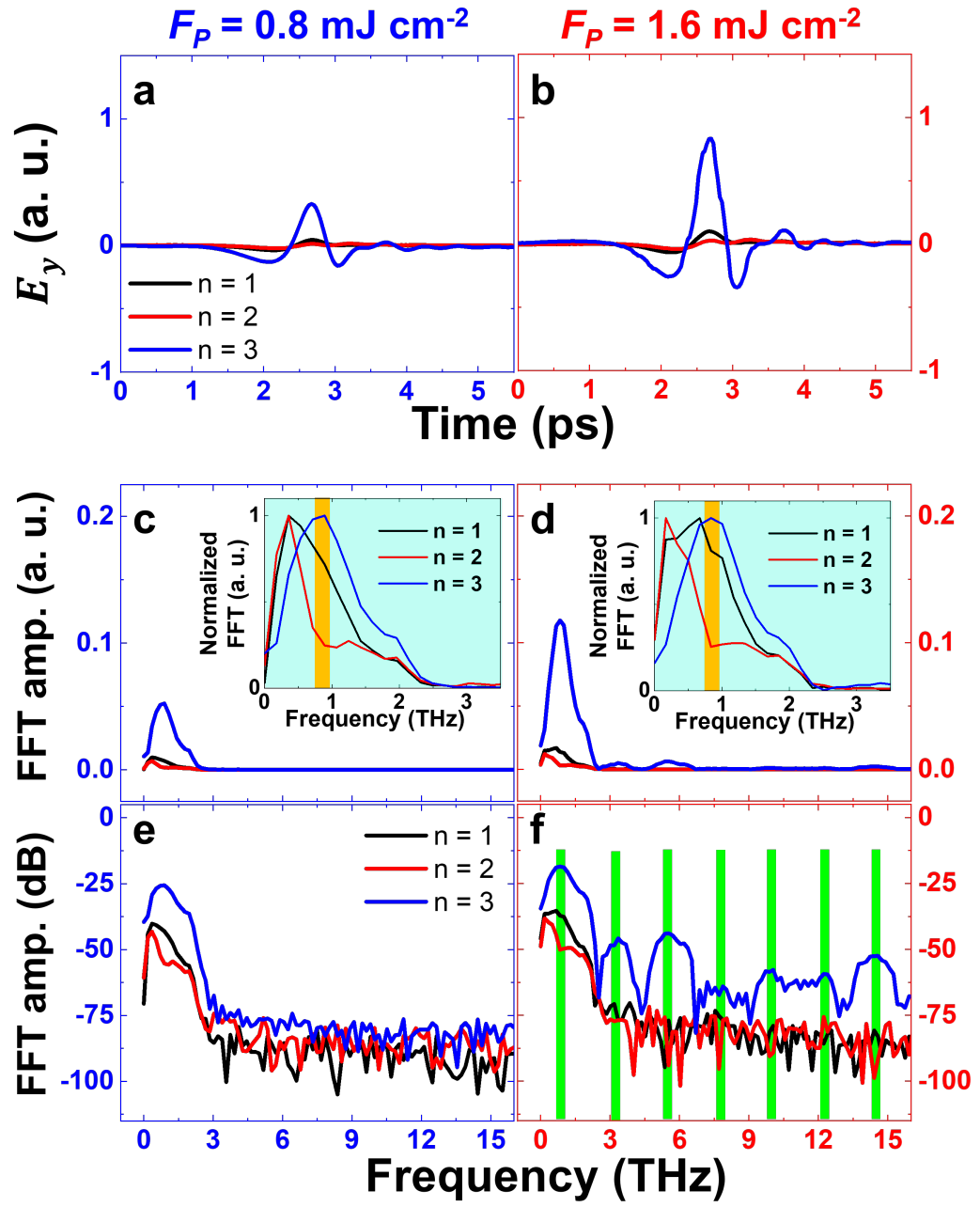


Fig. 2

[Pt/Co/W/MgO]_n

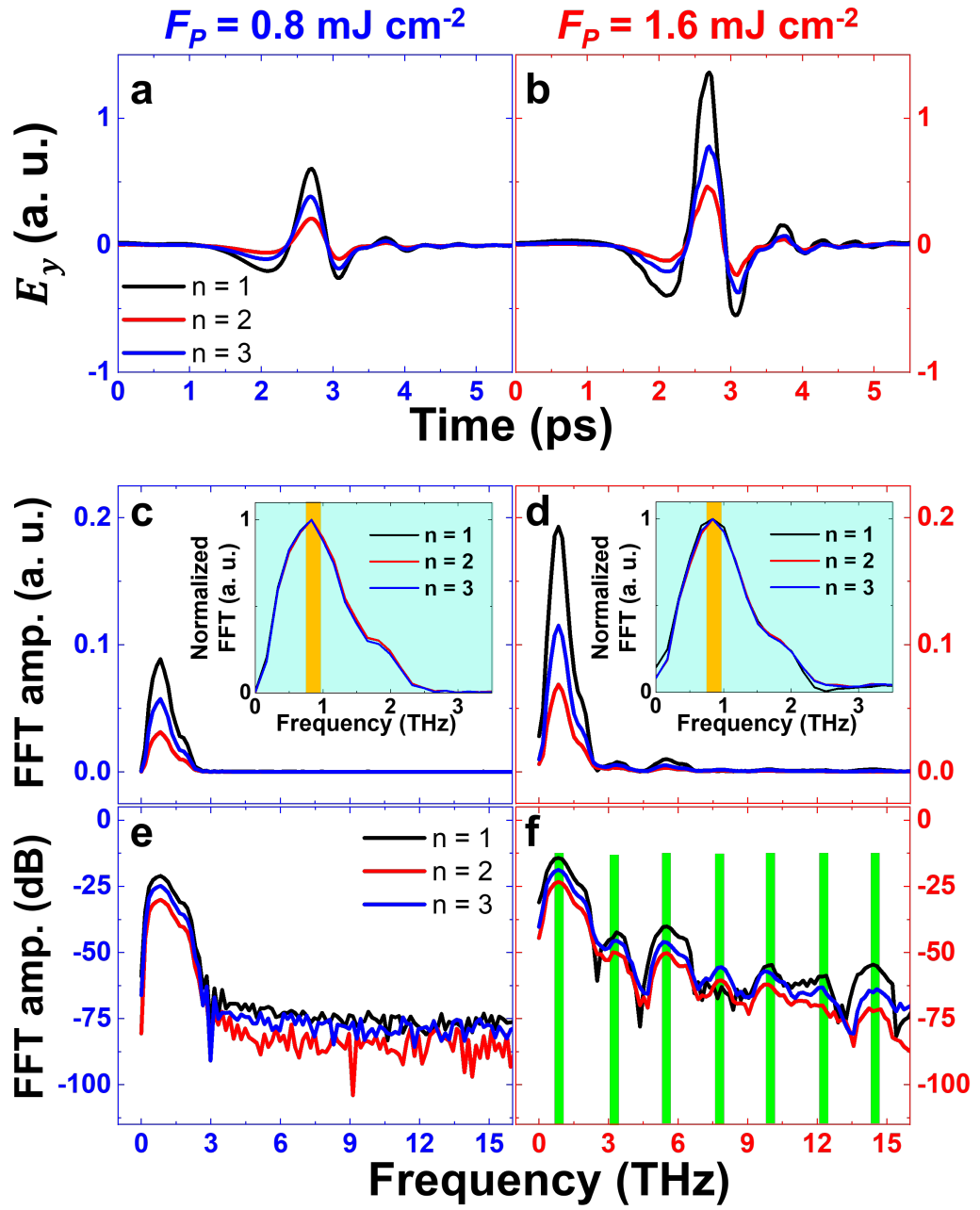


Fig. 3

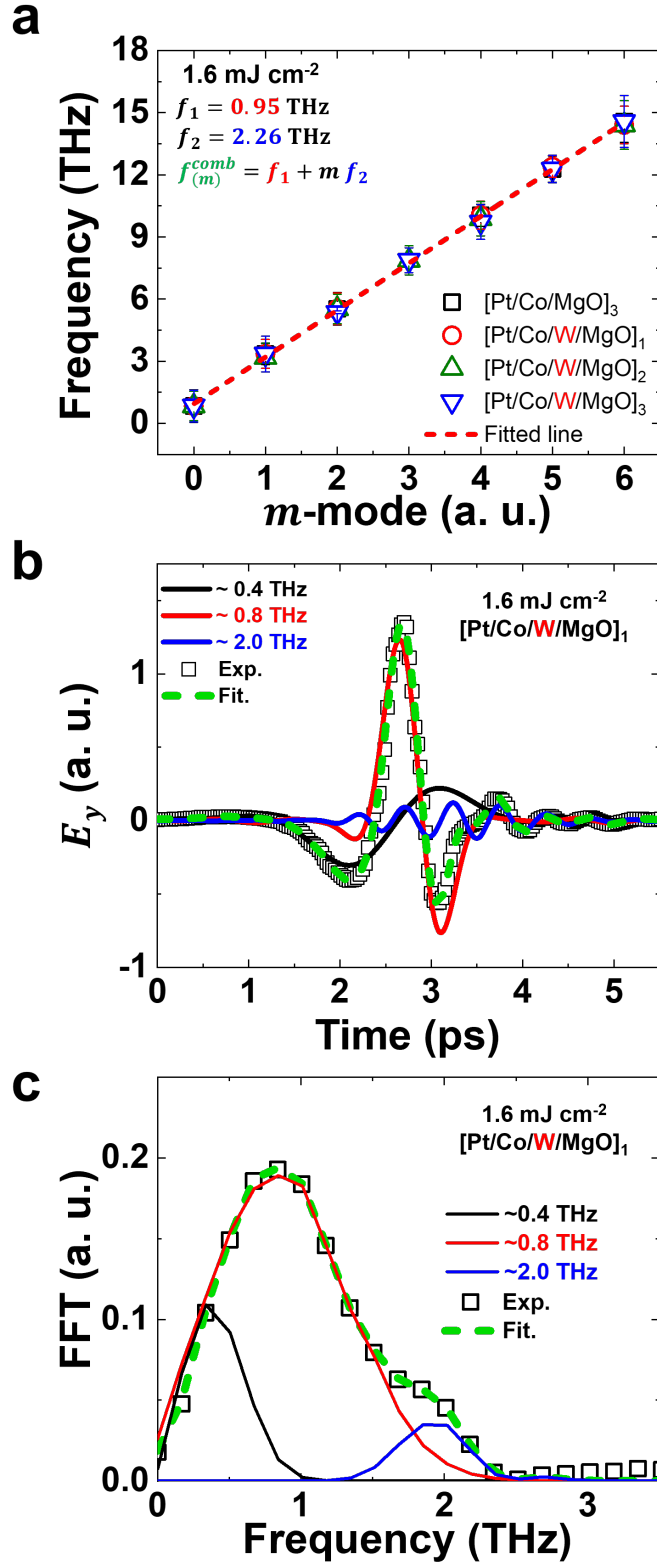


Fig. 4

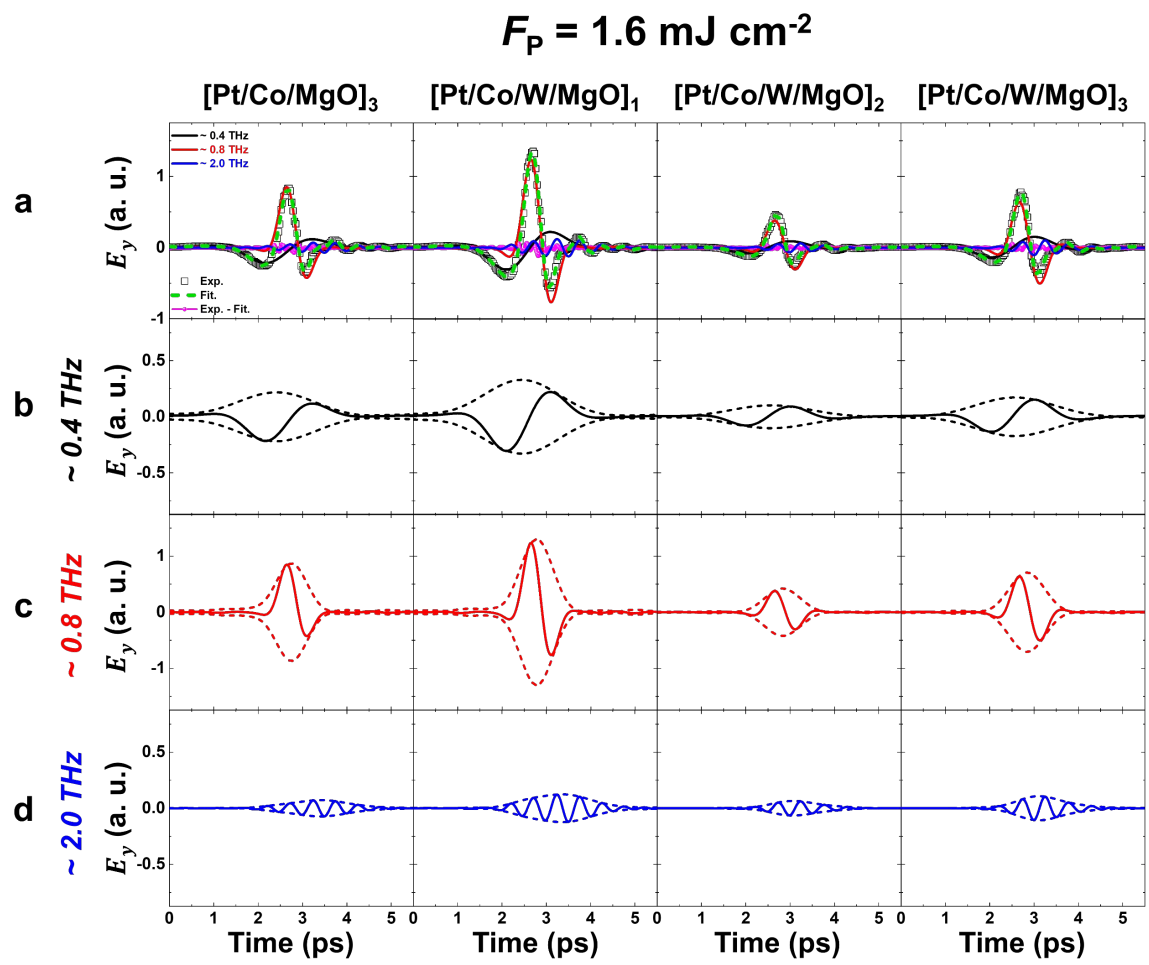


Fig. 5

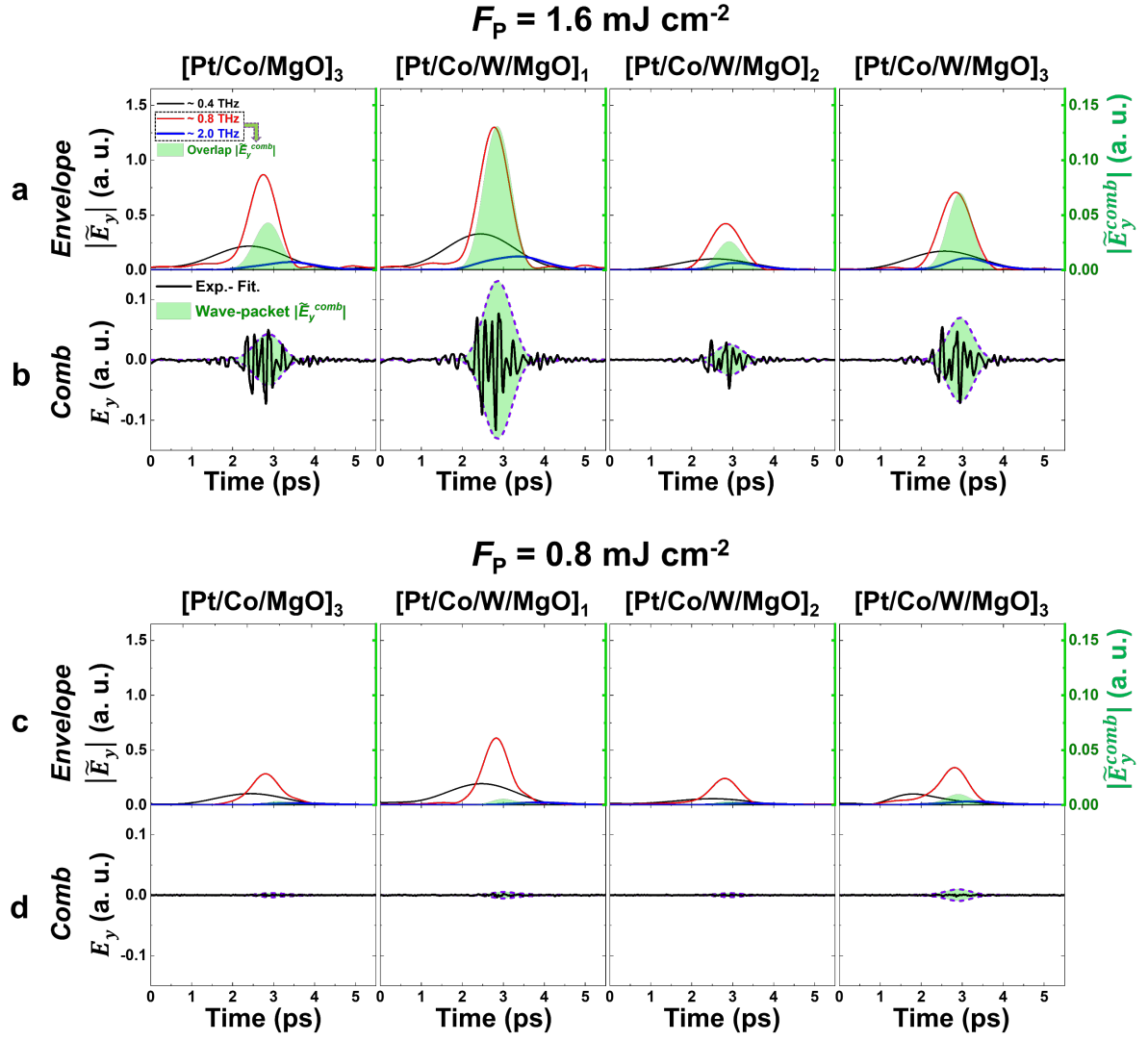


Fig. 6

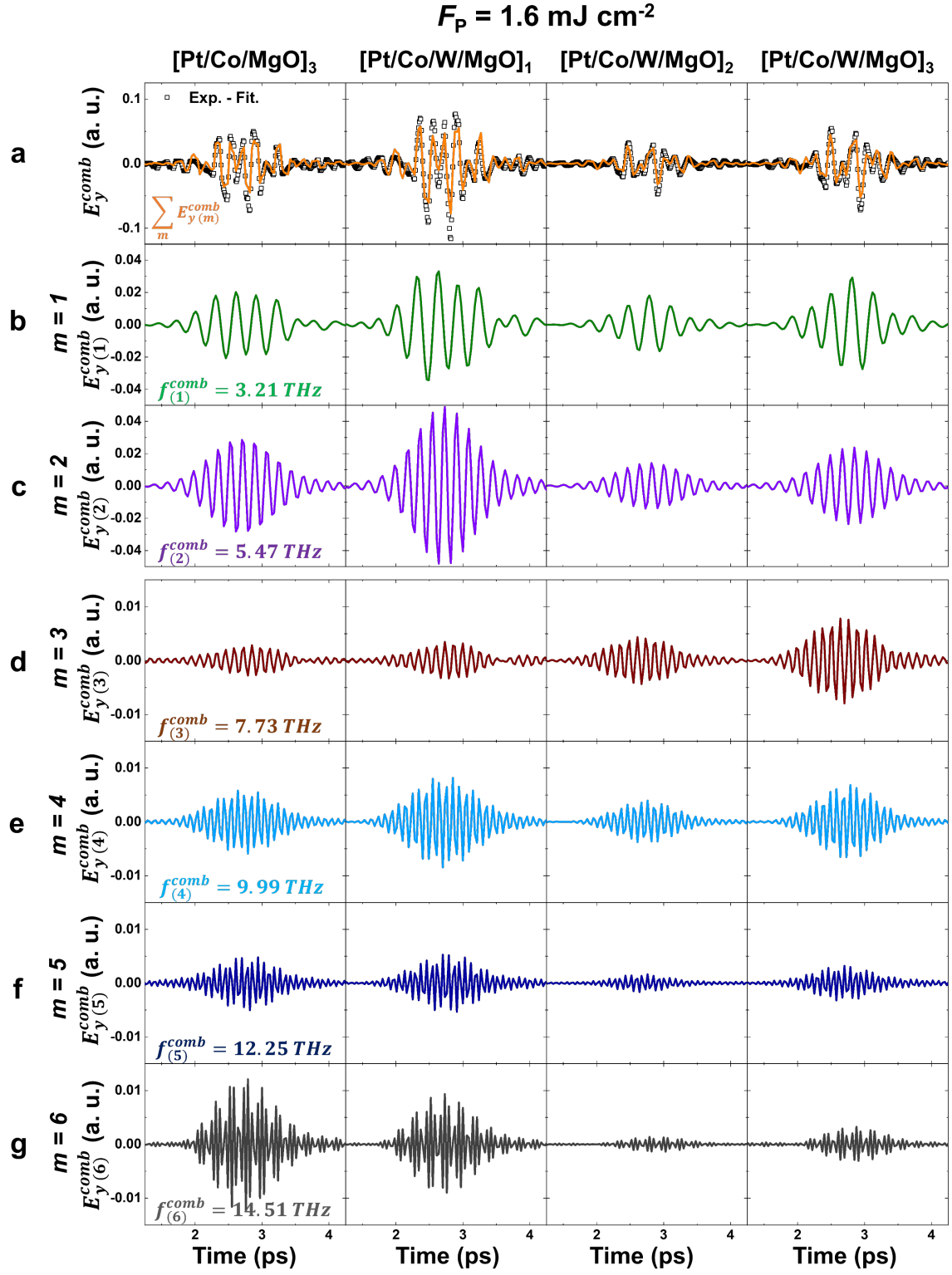


Fig. 7

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [SITHzcomborigin260505.docx](#)