Excitation and detection of coherent sub-terahertz magnons in ferromagnetic and antiferromagnetic heterostructures

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Excitation of coherent high-frequency magnons (quanta of spin waves) is critical to the development of high-speed magnonic devices. Here we computationally demonstrate the excitation of coherent sub-terahertz (THz) magnons in ferromagnetic (FM) and antiferromagnetic (AFM) thin films by a photoinduced picosecond acoustic pulse. Analytical calculations are also performed to reveal the magnon excitation mechanism. Through spin pumping and spin-charge conversion, these magnons can inject sub-THz charge current into an adjacent heavy-metal film which in turn emits electromagnetic (EM) waves. Using a dynamical phase-field model that considers the coupled dynamics of acoustic waves, spin waves, and EM waves, we show that the emitted EM wave retains the spectral information of all the sub-THz magnon modes and has a sufficiently large amplitude for near-field detection. These predictions indicate that the excitation and detection of sub-THz magnons can be realized in rationally designed FM or AFM thin-film heterostructures via ultrafast optical-pump THz-emission-probe spectroscopy.

INTRODUCTION

Ultrafast magnetoacoustics is focused on studying the interaction of femtosecond (fs)-laser-induced ultrashort (picosecond) acoustic pulse with magnetic order. It allows for investigating the magnetoelastic coupling at a possibly fastest attainable timescale and offers opportunities for the design of high-speed, compact, and energy-efficient magnetic devices. Due to the time compression of the laser energy, such photoinduced picosecond (ps) acoustic pulse can have an ultrahigh strain amplitude of >1% and short wavelengths down to a few nanometers (nm). In principle, such large and non-uniform strain would excite short-wavelength (high-frequency) magnons (quanta of spin waves) via magnetoelastic coupling, which underpin the development of high-frequency magnons (quanta of spin waves) via magnetoelastic coupling, which underpin the development of magnonic devices. Here we computationally demonstrate the excitation of coherent sub-terahertz (THz) magnons in ferromagnetic (FM) and antiferromagnetic (AFM) thin films by a photoinduced picosecond acoustic pulse. Analytical calculations are also performed to reveal the magnon excitation mechanism. Through spin pumping and spin-charge conversion, these magnons can inject sub-THz charge current into an adjacent heavy-metal film which in turn emits electromagnetic (EM) waves. Using a dynamical phase-field model that considers the coupled dynamics of acoustic waves, spin waves, and EM waves, we show that the emitted EM wave retains the spectral information of all the sub-THz magnon modes and has a sufficiently large amplitude for near-field detection. These predictions indicate that the excitation and detection of sub-THz magnons can be realized in rationally designed FM or AFM thin-film heterostructures via ultrafast optical-pump THz-emission-probe spectroscopy.

RESULTS

Excitation and detection of sub-THz magnons in ferromagnetic thin film

We propose a metal/dielectric/FM/HM thin-film heterostructure, as shown in Fig. 1a, using Al/MgAl2O4(001)/MgAl0.5Fe1.5O4(001)/Pt as an example. Recent computations predict that a ps acoustic pulse can excite multiple coherent standing magnon modes (n = 0, 1, 2, ... ∞) in a FM thin film via magnetoelastic coupling, and that the frequencies of high-order (n ≥ 1) magnon modes can reach sub-THz range. Polycrystalline Ni17 and (001) Fe81Ga19 thin films were utilized as examples. Here, we computationally demonstrate that such sub-THz magnon modes can be detected by adding a HM thin film on top of the FM thin film to enable the spin pumping at the FM/HM interface and spin-charge conversion in the HM layer via the inverse spin Hall effect (iSHE), and the emission of electromagnetic (EM) wave that arises mainly from the charge current. It is found that the emitted EM wave retains the spectral information of all the excited sub-THz magnons, providing a basis for magnon detection by THz emission spectroscopy.
been utilized to excite and detect fs-laser-induced THz spin current (incoherent magnons)\textsuperscript{22-26} and ultrafast demagnetization\textsuperscript{7,28}. It is worth noting that the addition of a spin-charge conversion layer is necessary for the detection of high-order magnon modes \((n \geq 1)\), because the net EM wave emission produced directly by the magnons via magnetic dipole radiation is negligible.

We now discuss the principles of materials selection for each layer of the heterostructure. First, polycrystalline Al film is commonly used as an optical-to-acoustic transducer\textsuperscript{7} for its large thermal expansion and small absorption length to near-infrared laser. Our previous computation\textsuperscript{19} predicts that tuning the Al film thickness \((\Delta t)\) of the MAFO thickness \((d)\) as a function of MAFO film thickness \((d)\). The frequencies for the case of \(d = 15 \text{ nm}\) are labeled.

An analytical theory on acoustic excitation of exchange-dominated standing magnons in isotropic FM thin films was presented in ref. \textsuperscript{17}, where the magneto-crystalline anisotropy was dropped. Here we derive an analytical expression of the magnon dispersion relation \(\omega(k)\) in FM thin films with cubic magneto-crystalline anisotropy (see Methods), where \(k\) is the angular wavevector along the film thickness direction and \(\omega\) is the angular frequency of the magnon. For standing magnons, \(k = \frac{2\pi mn}{d}\) \((n = 0, 1, 2, \ldots \infty)\) where \(d\) is the FM film thickness. This allows us to analytically calculate the frequency \(f = \frac{\omega}{2\pi[m]}\) of each magnon mode as a function of \(d\). Note that frequency of \(n = 0\) mode magnon is also the ferromagnetic resonance (FMR) frequency. The calculation results for \(001\) MAFO films are shown in Fig.\textsuperscript{1b}, which provide guidance on the acoustically mediated magnon excitation. For example, let us assume the MAFO film thickness is \(15 \text{ nm}\) and that the frequency window (illustrated as the vertical dashed line in Fig.\textsuperscript{1b}) contains up to \(500 \text{ GHz}\). In this case, three magnon modes, with frequency of \(422.8 \text{ GHz}\) \((n = 2)\), \(106.5 \text{ GHz}\) \((n = 1)\), and \(0.53 \text{ GHz}\) \((n = 0)\), can be excited.

To demonstrate this analytical prediction, we perform dynamical phase-field simulations (see Methods) to compute the spatiotemporal evolution of local magnetization in a 15-nm-thick \((001)\) MAFO film upon the injection of a bipolar Gaussian acoustic pulse, which has a peak amplitude \(\epsilon_{\text{max}} = 0.3\%\) in the adjacent \((001)\) MAO and a duration \(t = 6 \text{ ps}\), as indicated in Fig.\textsuperscript{1a}. Figure \textsuperscript{2a} presents the temporal profile of the \(\Delta m(t) = m_1(t)-m_2(t)\) at the top surface \((z = 15 \text{ nm})\) of the MAFO film in first \(0.5\) ns after the acoustic pulse propagates into the film from its bottom surface \((z = 0 \text{ nm})\), which shows the features of mixed high-frequency magnon modes and their amplitudes gradually decrease due to magnetic damping. The evolution of the \(\Delta m(t)\) at \(t = 2.5 \text{ ns}\) is plotted in Fig.\textsuperscript{2b}. At this stage, high-frequency magnon modes had attenuated, and a lower-frequency (ns-scale oscillation period), smaller-amplitude \((\sim 10^{-3})\) magnon mode can be seen. Figure \textsuperscript{2c} shows the frequency spectra of both the \(\Delta m_1, \Delta m_2\) within \(t = 0-8.5 \text{ ns}\) and the strain averaged over the MAFO thickness \(<\epsilon_z, \epsilon_z\>\). As shown, three distinct magnon modes \((m = 0, 1\) and \(2)\) are excited and their frequency values match the analytical calculation almost exactly. Higher-mode magnons, e.g., \(950 \text{ GHz}\) for \(n = 3\) mode, were not excited, because their frequency values fall outside the frequency window of the acoustic pulse \((0-600 \text{ GHz})\). More detailed analyses indicate that the amplitudes of the magnons are proportional to the spectral amplitude of the acoustic pulse at the corresponding frequency (see Supplementary Note 1). For further demonstration, we extracted the spatial profiles of the three magnon modes by performing inverse Fourier transform of the spectrum \(\Delta m_1(\omega)\) over the entire MAFO film with all non-peak-frequency components filtered out. As shown in Fig.\textsuperscript{2d}, the obtained profiles display canonical features of the \(n = 0, 1,\) and \(2\) magnon modes (c.f., the schematics in Fig.\textsuperscript{1a}).
For detection, we suggest that the spectral features of these magnon modes should be retained in the frequency spectra of the charge current $J$ in the adjacent Pt layer as well as the EM wave emitted by the $J$. This is because $J$ arise from the precessing local magnetization at the MAFO/Pt interface $\Delta m(z = 15 \text{ nm}, t)$, and because the frequency spectrum of $\Delta m(z = 15 \text{ nm}, t)$ (Fig. 2c) contains all the excited magnon modes. Figure 3a shows the spatiotemporal profile of the finite charge current density $J_z(t)$ in the Pt layer, which is a sum of the current generated via the iSHE $J^{\text{iSHE}}$ and the eddy current (polarization current) $J^{p}$ induced by the emitted EM wave (see details in Methods). As seen, the $J_z$ in the half thickness of the Pt layer is opposite to that in the other half. This is because (1) the directions of $J^{\text{iSHE}}$ and $J^{p}$ are opposite to each other; (2) the amplitude of $J^{\text{iSHE}}$ decreases monotonically along the Pt thickness direction while $J^{p}$ is almost spatially uniform in the Pt layer due to the long wavelength (millimeter-scale) of the emitted EM wave. Distributions of $J^{\text{iSHE}}$ and $J^{p}$ are shown in Supplementary Fig. 2. Figure 3b shows the temporal evolution of the electric-field component of the emitted EM wave $E_x(t)$ in the free space (at 5 nm above the Pt top surface), which decreases over time as the magnon amplitude decreases (c.f. Fig. 2a). The peak amplitude of the $E_x(t)$, ~80 V/m, is large enough for detection by the time-domain electro-optical sampling used in ultrafast THz emission spectroscopy (e.g., see refs. 22,23). Figure 3c shows the frequency spectrum of the $E_x(t)$. Two discrete peak frequencies can be seen, which have the same values as those of $n = 1$ and 2 mode magnon (c.f. Fig. 2c). Notably, although the spectral amplitude of the $n = 2$ mode magnon is smaller than that of the $n = 1$ mode magnon (Fig. 2c), the spectral amplitude of the EM wave contributed by the $n = 2$ mode magnon is larger because higher-frequency magnons pump larger-amplitude spin current into the Pt (see Methods). The EM wave emission from the $n = 0$ mode magnon (FMR) is negligible because of its small amplitude (Fig. 2b) and low frequency (Fig. 2c). Furthermore, the emitted EM wave is circularly polarized, as shown in Fig. 3d, due to the phase difference between $E_x$ and $E_y$.

**Excitation and detection of sub-THz magnons in antiferromagnetic thin film**

We now show that coherent standing magnon modes can likewise be excited in an AFM thin film by a ps acoustic pulse via magnetoelastic coupling. A similar metal/dielectric/AFM/HM heterostructure is considered (Fig. 4a), using Al/MgO/(001)/FeMn$_{50}$/Pt as an example. Polycrystalline FeMn$_{50}$/Pt$_{50}$ (FeMn) film is considered as the representative AFM material due to its robust magnetoelastic coupling ($B_s = 9.7$ MJ m$^{-3}$)33,34. FeMn can be modeled as an easy-axis antiferromagnet with two magnetic sublattices33,35 whose magnetizations are denoted as $m^{(1)}$ and $m^{(2)}$. Following the same approach used for the FM film, an analytical formulation of the magnon dispersion relation $\omega(k)$ is derived for easy-axis AFM thin films (see Methods). Figure 4b shows the analytically calculated frequency $f = \omega/2\pi$ of each AFM magnon mode as a function of the FeMn film thickness $d$. Comparing Figs. 1b and 4b, it can be seen that the antiferromagnetic resonance (AFMR) frequency of the $n = 0$ mode AFM magnon, is much higher than the FMR frequency. Considering a 15-nm-thick FeMn film and the frequency window of the injected acoustic pulse reaches up to 200 GHz (illustrated as the vertical dashed line in Fig. 4b), three AFM magnon modes, with frequency of 192.6 GHz ($n = 2$), 91.3 GHz ($n = 1$), and 49.3 GHz ($n = 0$), can be excited. The dynamical phase-field modeling results demonstrating this analytical prediction are shown in Fig. 5, where the evolution of both the $\Delta m^{(1)}$ and $\Delta m^{(2)}$ (see Fig. 5a) were utilized to analyze the frequency spectra of the AFM magnon modes (Fig. 5b). Moreover, as shown in Fig. 5a, the $\Delta m^{(1)}$ and $\Delta m^{(2)}$ are opposite to each other during the evolution, and it is noteworthy that the $m^{(1)}$ and $m^{(2)}$ are precessing counterclockwise and clockwise around the [111] easy axis, respectively, as sketched in the inset of Fig. 5a.

The precession of both the $m^{(1)}$ and $m^{(2)}$ at the FeMn/Pt interface can pump spin currents into the Pt layer. Principles of spin pumping from an easy-axis antiferromagnet have been discussed elsewhere16–18. Figure 6a shows the spatiotemporal profile of the total charge current density $\mathbf{J}$ in both FeMn and Pt films. The $\mathbf{J}$ is larger in Pt because the $\mathbf{J}^{\text{iSHE}}$ exists only in the Pt layer. Figure 6b presents the temporal evolution of the in-plane electric-field component $E_x(t)$ of the emitted EM wave in the free space (5 nm above the Pt top surface). The amplitude of $E_x(t)$ decreases over time as the magnon amplitude decreases (c.f., Fig. 5a). The frequency spectrum of the $E_x(t)$ in Fig. 6c reveals three distinct peak frequencies, which have same values as the excited AFM magnon modes $n = 0$, 1, and 2 (c.f., Figs. 4b and 5b). Moreover, the emitted EM wave is linearly polarized with no phase difference between $E_x$ and $E_y$, as shown in Fig. 6d (see detailed explanation in Methods section).
heavy metal (HM) thin film to enable spin pumping and spin-charge conversion. Specifically, since the frequency spectra of the magnon modes are similar to the frequency spectra of the charge current in the HM layer and the free-space electromagnetic (EM) wave emission, the excited magnons can be detected by measuring the EM wave emission. Compared to the commonly used method of ultrafast TR-MOKE where the signals of sub-THz magnons (which have nm-scale wavelength) can be averaged out due to the nm-scale penetration depth of the probe depths, detecting the EM wave emission allows for retaining the spectral information of all magnon modes.

In order to computationally demonstrate the proposed principles of detection and accurately model the emitted EM wave, we have developed an in-house dynamical phase-field model that considers fully coupled dynamics of acoustic waves, spin waves, and EM waves, which has previously not been considered together despite a few advanced computational models in this regard. The physical validity and high numerical accuracy of our phase-field model can be seen from the almost exact match between the analytically calculated and simulated frequencies of the magnon modes (Figs. 2c and 5b).

Results on the validation of other modules, especially our in-house finite-difference time-domain (FDTD) solver for EM wave generation and propagation, can be found in Supplementary Note 2 and Supplementary Figs. 3–5. The predicted peak amplitude of the emitted electric field, on the order of 100 V/m, is sufficiently large for detection. The emission mainly arises from the charge current in the HM layer via electric dipole radiation, because the net charge current in the HM layer and the free-space electromagnetic (EM) wave emission, the excited magnons can be detected by measuring the EM wave emission. Compared to the commonly used method of ultrafast TR-MOKE where the signals of sub-THz magnons (which have nm-scale wavelength) can be averaged out due to the nm-scale penetration depth of the probe depths, detecting the EM wave emission allows for retaining the spectral information of all magnon modes.

**Fig. 3 Magnon detection.** a Spatiotemporal profile of the total charge current density $J_\gamma^c$ in the 7-nm-thick Pt. b Temporal profile of the emitted electric-field component $E_x(t)$ at 5 nm above the Pt top surface within $t = 0–200$ ps and c its frequency spectrum. d Evolution of the emitted electric field vector $\mathbf{E}(t)$ within $t = 0–15$ ps, showing a circular polarization.

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**METHODS**

**Dynamical phase-field model**

Below we describe the individual modules for spin wave (magnon) dynamics, acoustic wave (coherent phonons), and EM wave (photon) dynamics, and how these modules are coupled together in a multilayer heterostructure. Here the ‘coupled’ means bidirectional phonon-magnon and magnon-photon coupling. Specifically, the acoustically excited local magnetization will generate secondary acoustic waves via magnetoelastic backaction. The EM wave, which originates from the precession of magnetic dipoles (via spin-charge conversion), will in turn affect the magnetization dynamics via its magnetic-field component. Both types of back-actions, despite being weak in the present cases, have been incorporated. Moreover, our model is GPU (Graphics Processing Unit) accelerated to facilitate high-throughput modeling and heterostructure design.

**Part 1: Spin wave dynamics**

The simulations were performed with a dynamical phase-field model that considers the fully coupled dynamics of elastic waves (acoustic phonons), spin waves (magnons), and EM waves in a heterostructure with discontinuous magnetic and elastic properties across the interface. The evolution of normalized local magnetization $\mathbf{m}$ in a ferromagnetic (FM) system and $\mathbf{m}^{\text{AFM}}$ ($s = 1,2$) in two magnetic sublattices of an antiferromagnetic (AFM) system are governed by the LLG equation. The LLG equation for the FM system is expressed as,

$$\dot{\mathbf{m}} = -\gamma \frac{\mathbf{m} \times \mathbf{H}_{\text{eff}}}{\alpha} - \frac{\alpha}{\gamma} \mathbf{m} \times (\mathbf{m} \times \mathbf{H}_{\text{eff}})$$

(1)

where $\gamma$ is the gyromagnetic ratio; $\alpha$ is the magnetic damping coefficient. The total effective magnetic field $\mathbf{H}_{\text{eff}} = \mathbf{H}^{\text{mag}} + \mathbf{H}^{\text{exch}} + \mathbf{H}^{\text{anis}} + \mathbf{H}^{\text{em}}$. Among them, the magnetocrystalline anisotropy field $\mathbf{H}^{\text{anis}}$ is given by,

$$\mathbf{H}^{\text{anis}} = -\frac{1}{\mu_0 C_1} \left[ K_1 \left( m_x^2 + m_y^2 \right) + K_2 m_y^2 m_z^2 \right] \mathbf{m}.$$  

(2)
where $\mu_0$ is vacuum permeability; $M_s$ is saturation magnetization; $K_{1}$ and $K_{2}$ are magnetocrystalline anisotropy coefficients; $i = x, y, z$, and $j \neq i, k \neq i, j$. The magnetic dipolar coupling field $H^{\text{exch}} = \frac{\mu_0 M_s}{4\pi} \mathbf{V} \cdot \mathbf{m}$ describes the back-action of the EM wave on local magnetization $\mathbf{m}$ on the local magnetization dynamics, written as,

$$H^{\text{exch}} = \frac{\mu_0 M_s}{4\pi} \mathbf{V} \cdot \mathbf{m},$$

where $B_1$ and $B_2$ are magnetoelectric coupling coefficients; $i = x, y, z$, and $j \neq i, k \neq i, j$. The magnetic dipolar coupling field $H^{\text{exch}} = (0, 0, -M_s m_0)$ is applied along the $z$ axis to lift magnetizations off the $xy$ plane by 45° before acoustic excitation, so that the torque exerted by the $H^{\text{exch}}$ on the magnetizations is maximized. The magnetic field component of the emitted EM wave $H^{\text{EM}}$ describes the back-action of the EM wave on local magnetization dynamics. The calculation of $H^{\text{EM}}$ will be detailed later.

The LLG equations for the two magnetic sublattices of the AFM system have the same form as the FM but with their own total effective field $H^{\text{eff}}$ (s = 1, 2).

$$\frac{3m^{(s)}_{i,j}}{\alpha m} = -\frac{1}{\mu_0 M_s} \left( B_1 m^{(s)}_{i,k} + B_2 \left( m^{(s)}_{i,j} + m^{(s)}_{k,j} \right) \right),$$

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In this work, the gyromagnetic ratio $\gamma$ and magnetic damping coefficient $\alpha$ are set to be same for both sublattices as approximation, following refs. 33,35. The total effective magnetic field $H^{\text{eff}} = H^{\text{mexch}} + H^{\text{exch}} + H^{\text{dip}} + H^{\text{EM}}$ describes the back-action of the EM wave on local magnetization dynamics. The model considers an easy-axis AFM system for which the magnetocrystalline anisotropy fields $H^{\text{mexch}}$ in both sublattices are given by,

$$H^{\text{mexch}}(i) = \frac{2K_{1s}}{\mu_0 M_s} \mathbf{a}_i \cdot \mathbf{m}^{(s)} \mathbf{a}_i,$$

where $K_{1s}$ is uniaxial anisotropy coefficient; $\mathbf{a}_i$ is unit vector in the direction of easy axis; $K_{1s}$ and $M_s$ are assumed to be same for the two sublattices. For illustration, $\mathbf{a}_i$ is set to be along [111] in this work and the $m^{(1)}$ and $m^{(2)}$ are directed along [111] and [TTT] before acoustic excitation, respectively. The principles of magnon excitation and detection are independent of the easy-axis orientation. The intra-lattice magnetic exchange coupling field $H^{\text{exch}} = \frac{\mu_0 M_s}{4\pi} \mathbf{V} \cdot \mathbf{m}$ has the same form as that in the AFM system and $A_{n}$ is assumed to be same for two sublattices. The magnetic boundary condition $\partial m^{(s)} / \partial z = 0$ is applied on the two surfaces of the AFM film. The magnetoelastic field is in hand in magnitude for AFM in comparison to the form for FM (Eq. (3)) to ensure that saturation magnetostriction $A_{n}$ occurs when $m^{(1)}$ and $m^{(2)}$ are coaxially directed $33$,

$$H^{\text{exch}} = \frac{1}{\mu_0 M_s} \left( B_1 m^{(s)}_{i,k} + B_2 \left( m^{(s)}_{i,j} + m^{(s)}_{k,j} \right) \right),$$

where magnetoelastic coupling effects in both sublattices are assumed to be same. Therefore, the same set of $B_1$ and $B_2$ is used for both sublattices; $i = x, y, z$, and $j \neq i, k \neq i, j$. It is noteworthy that $B_1 = B_2$ for magnets with isotropic elasticity. The two sublattices also share the same magnetic dipolar coupling field $H^{\text{exch}} = (0, 0, -M_s m^{(1)} - M_s m^{(2)})$ which includes contribution from both sublattices. Moreover, we set $H^{\text{exch}} = 0$. Before acoustic excitation, the AFM film is set to be a single AFM domain, that is, the Néel vector $\mathbf{n} = 0.5(m^{(1)} + m^{(2)})$ is spatially uniform and along the [111]
easy axis. Such single AFM domain could be obtained by cooling the magnet from its high-temperature paramagnetic phase in the presence of a bias magnetic field applied along the easy axis. After then, the bias magnetic field can be removed. Different from the case of FM film, a single AFM domain can remain stable at $H_{\text{eff}} = 0$. The effective field produced by the inter-lattice AFM-type exchange coupling favors opposite alignment of $m^1$ and $m^2$, calculated as,

$$H_{\text{AFM}}^{(1)} = -J_{\text{AFM}}^m m^2$$

$$H_{\text{AFM}}^{(2)} = -J_{\text{AFM}}^m m^1$$

where $J$ is the AFM exchange coupling coefficient. The sublattices share the same $H_{\text{EM}}$ from EM wave.

**Part 2: EM dynamics in magnet/heavy-metal heterostructure**

To calculate the EM wave dynamics of electric and magnetic dipole radiation, an in-house FDTD solver of Maxwell's equations was developed. The two governing equations are listed below:

$$V \times E_{\text{EM}} = -\mu_0 \left( \frac{\partial E_{\text{EM}}}{\partial t} + \frac{\partial M}{\partial t} \right)$$

$$V \times H_{\text{EM}} = \sigma E_{\text{EM}} + J^p + J^s$$

where $E_{\text{EM}}$ is the electric field component of the EM wave for the main results; $M$ is the local magnetization. The temporally changing $M$ provides the source of magnetic dipole radiation. For FM system, $M = M_0 m$, where $m$ can be obtained by solving the LLG equation (Eq. (1)). For AFM system, $M = M_0^1 m^1 + M_0^2 (m_0^1 + m_0^2)$ (assuming that $M_0$ is the same in both sublattices), where $m_0^1$ and $m_0^2$ are likewise obtained by solving LLG equations (Eq. (4)). The LLG and Maxwell's equations (Eqs. (9) and (10)) are coupled and solved simultaneously. $D$ is electric displacement field; $J^p$ is polarization current induced by the electric field $E_{\text{EM}}$ in dispersive medium, which causes the absorption and reflection of the EM wave. The polarization current $J^p$ (or eddy current) in metallic conductors (Pt and FeMn in this work) is obtained by solving time-domain auxiliary differential equation (ADE) based on Drude model,

$$\frac{\partial J^p}{\partial t} + \tau^{-1} J^p = \lambda_\text{sd} \omega_e^2 E_{\text{EM}}^2$$

where $\lambda_\text{sd}$ and $\tau$ denote the plasma frequency and electron relaxation time, respectively. For alloy such as Fe$_x$Mn$_{100-x}$, the total $J^p$ is treated as the composition-weighted average of the $J^p$ of each metallic component, i.e., $0.5 J^p_{\text{Fe}} + 0.5 J^p_{\text{Mn}}$. This ensures that the frequency-dependent relative permittivity $\varepsilon_r(\omega)$ of the alloy is also the composition-weighted average of the $\varepsilon_r(\omega)$ of constituent metallic components. The ADE (Eq. (11)) and the Maxwell's equations (Eqs. (9) and (10)) are coupled and solved simultaneously. The $J^s$ is the free charge current density. In this work, $J^s$ is converted from the spin current density $J^s$ via the iSHE ($J^s = J^\text{iSHE}$) and is the source of electric dipole radiation.

The spin current density $J^s(t)$ from the AFM spin pumping is calculated as the sum of contribution from both sublattices via the relation, $J^s(t) = \sum \text{Re} \{ \frac{1}{\lambda} Re[\mu_{\text{sd}}(\omega)] m^{(1)} + m^{(2)} \} \times \frac{\partial \mu_{\text{sd}}}{\partial \omega} \phi$, where $\mu_{\text{sd}}$ is the inter-lattice AFM-type exchange coupling favors opposite alignment of $m^{(1)}$ and $m^{(2)}$ calculated as,

$$\frac{\partial J^p}{\partial t} + \frac{\partial J^s}{\partial t} = \lambda_\text{sd} \omega_e^2 E_{\text{EM}}^2$$

where $\lambda_\text{sd}$ and $\tau$ denote the plasma frequency and electron relaxation time, respectively. For alloy such as Fe$_x$Mn$_{100-x}$, the total $J^s$ is treated as the composition-weighted average of the $J^s$ of each metallic component, i.e., $0.5 J^s_{\text{Fe}} + 0.5 J^s_{\text{Mn}}$. This ensures that the frequency-dependent relative permittivity $\varepsilon_r(\omega)$ of the alloy is also the composition-weighted average of the $\varepsilon_r(\omega)$ of constituent metallic components. The ADE (Eq. (11)) and the Maxwell's equations (Eqs. (9) and (10)) are coupled and solved simultaneously. The $J^s$ is the free charge current density. In this work, $J^s$ is converted from the spin current density $J^s$ via the iSHE ($J^s = J^\text{iSHE}$) and is the source of electric dipole radiation.

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Part 3: Acoustic wave dynamics in multilayer

The evolution of mechanical displacement $u$ is obtained by solving an elastodynamic equation which incorporates the magnetostrictive stress to describe the backaction of magnetoelasticity on elastic wave dynamics, $\rho\ddot{u} = V \cdot (\mathbf{e} - \mathbf{e}^s)$, where $\rho$ and $c$ are phase-dependent mass density and elastic stiffness, respectively. A full tensorial expansion of this elastodynamic equation is provided in our previous work. $\mathbf{e}$ is related to $u$ via $e_i = \left(\frac{\partial u_i}{\partial x_j} + \frac{\delta_{ij}}{2}\right)$, $i, j = x, y, z$. $\mathbf{e}^s$ is the eigenstrain produced by magnetization via magnetostriction. For a FM system with a cubic parent phase, $\mathbf{e}^s$ takes the conventional form of

$$e_i^s = \frac{\lambda_{FM}}{V_{000}} (m_{i0}^2 - \frac{1}{3}) + \frac{\lambda_{FM}}{V_{111}} m_{ij} m_{ji},$$

(12)

where $\lambda_{100}$ and $\lambda_{111}$ are saturation magnetostriiction along the local $<100>$ and $<111>$ axes. For an isotropic AFM system, one has $\lambda_{FM}$

$$e_i^s = \lambda_{AFM} (m_{i1}^2 - \frac{1}{3}) + \lambda_{AFM} (m_{i2}^2 - \frac{1}{4}),$$

(13)

where $\lambda_{AFM}$ is the magnetostrictive coefficient. Note that $i = x, y, z, j \neq i$. The LLG equations (Eqs. (1) and (4)) and the elastodynamic equation are coupled and solved simultaneously. For simplicity, the injection of the ultrashort acoustic pulse is modelled by applying a time-dependent mechanical displacement at the AI/MAO or AI/MgO interface in the form of a Gaussian function, $u(t) = u_{max} \exp(-t^2/\sigma^2)$, which leads to a bipolar longitudinal strain $\varepsilon_{Lz} = \partial u_{Lz}/\partial z$ propagating in the dielectric layer (MAO or MgO) and then the magnetic layer (MAO or FeMn), as sketched in Figs. 1a and 4a. Such bipolar Gaussian acoustic pulse is a commonly used approximation for describing fs-laser-induced ultrafast acoustic pulse from polycrystalline metal transducer such as AI/MAO. The dielectric layer (MAO or MgO) is set to be sufficiently thick (e.g., hundreds of micrometers) to serve as a perfect sink of the reflected acoustic pulse. The entire heterostructure is discretized into one-dimensional (1D) computational cells along $z$ direction, with cell size $\Delta z = 0.2$ nm. Central finite difference is used for calculating spatial derivatives. All equations are solved simultaneously using the classical Runge-Kutta method for time-marching with a real-time step $\Delta t = 5 \times 10^{-15}$ s.

The materials parameters are summarized below. For (001) MAO, the elastic stiffness coefficients $c_{11} = 282.9$ GPa, $c_{12} = 155.4$ GPa, $c_{44} = 154.8$ GPa and mass density $\rho = 3578$ kg m$^{-3}$. For (001) MAO thin film, the elastic stiffness coefficients are assumed to be the same as MAO: $\rho = 4355$ kg m$^{-3}$, gyromagnetic ratio $\gamma = 0.227 \text{ rad MHz}^{-1}$ m$^{-1}$; the damping coefficient $\alpha = 0.021 \text{ at} \theta = 0.015$ is the intrinsic Gilbert damping coefficient without spin pumping; $\alpha' = \alpha_{FM} + \alpha_{AFM}$ is the magnetic damping induced by spin pumping (65) ($\alpha_{FM}$ is the g-factor, $\mu_0$ is the Bohr magneton); saturation magnetization $M_s = 0.0955$ MA m$^{-1}$; the exchange coupling coefficient $A_{FM}$ is assumed to be the same as CoFe$_2$O$_4$; magnetoelastic anisotropy coefficient $K_{\text{FM}} = -477.5$ J m$^{-1}$, magnetoelectric coupling coefficient $B_1 = 1.2$ MJ m$^{-3}$ and $B_2 = 0$. For MgO, $c_{11} = 297$ GPa, $c_{12} = 95.9$ GPa, $c_{44} = 156$ GPa and $\rho = 3580$ kg m$^{-3}$. For polycrystalline FeMn with isotropic elasticity properties, $c_{11} = 103.7$ GPa, $c_{12} = 44.4$ GPa, $c_{44} = 29.6$ GPa, which are calculated based on Young’s modulus of 77 GPa and Poisson’s ratio of 0.3 and assumption of isotropic elasticity; $\rho = 7700$ kg m$^{-3}$; $\gamma = 0.221$ rad MHz$^{-1}$ m$^{-1}$; the intrinsic Gilbert damping coefficient $\alpha = 0.0045$ is the intrinsic Gilbert damping coefficient of FeMn (67) (some of these parameters are assumed to be the same as the Fe-$\text{Mn}$ for simplicity). The uniaxial anisotropy coefficient $K_{A} = M_{s}$ is assumed to be $5 \times 10^5$ J m$^{-3}$. The coefficient for AFM-type exchange coupling $J = 3.97 \times 10^4$ J m$^{-3}$. The magnetostatic coefficients $H_{\text{AFM}} = 109$ ppm based on a recent experiment (68), hence $B_1 = B_2 = -1.5 \times 10^7$ GPa, $c_{11} - c_{12} = -3 \times 10^4 \text{ Wb/m}^2$, $-9.7$ MJ m$^{-3}$. For Pt/Fe, $c_{11} = 347$ GPa, $c_{12} = 250$ GPa and $\rho = 21,450$ kg m$^{-3}$ (some of these parameters are assumed to be the same as the Fe-$\text{Mn}$ for simplicity). The uniaxial anisotropy coefficient $K_{A} = M_{s}$ is assumed to be $5 \times 10^5$ J m$^{-3}$.
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