Optical-acoustic excitation of broadband terahertz antiferromagnetic spin waves

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Abstract

We propose an optical-acoustic means to excite broadband terahertz antiferromagnetic (AFM) spin wave in a metal/insulator/antiferromagnet heterostructure. The AFM spin wave is excited by an ultrafast strain wave triggered by a femtosecond pulsed laser based on photoacoustic conversion. This spin wave comprises an AFM exchange spin wave and a magnetoelastic spin wave. Their dispersion curves are overlapped in a wide frequency range by manipulating the Dzyaloshinskii–Moriya interaction, which is accompanied by lifting the degeneration of the spin-wave modes with opposite chirality. This optical-acoustic excitation of spin waves exploits the laser-induced ultrafast strain waves and avoids the thermal effect from the laser. It paves a way to develop novel AFM devices that can apply for ultrafast information processing and communication.

1. Introduction

Terahertz (THz) radiation (electromagnetic (EM) radiation with frequency ranging from 0.3 to 3 THz) attracts extensive interest owing to its unique properties and extraordinary potential applications \cite{1}. For example, THz waves can penetrate an optically opaque medium with a great signal-noise ratio, valuable in high-resolution imaging, wireless communication, and security check \cite{2}. THz waves are also safe in biomedical applications due to low photon energy \cite{3}.

A strong antiferromagnetic (AFM) exchange field sustains THz magnetization precession in an AFM medium, generating THz EM waves \cite{4}. This THz precession is triggered by the interaction between the EM field of a femtosecond (fs) laser and the AFM moments \cite{4–8}. In addition to a uniform AFM precession, Hortensius \textit{et al} also realized generation and detection of a coherent broadband AFM spin-wave propagation excited by an ultrashort pulse of laser \cite{9}. However, the laser transmission in an optically transparent AFM medium may bring about a thermal effect that weakens the exchange coupling \cite{10}. This is especially problematic for a metallic AFM medium. In recent years, efforts have been paid to non-thermal excitation of ultrafast magnetization precession \cite{11}, such as ultrafast AFM dynamics via light-driven phonons \cite{12} and ultrafast manipulation of magnetic parameters by photo-electron interaction \cite{13, 14}.

In addition to laser, a picosecond (ps) strain wave may also induce THz magnetization dynamics \cite{15}. Barra \textit{et al} have predicted THz AFM dynamics induced by a transient ps strain wave based on the inverse piezoelectric effect via switching on a DC voltage source \cite{16}. This technique avoids the thermal effect from the laser but cannot trigger continuous THz emission.
Figure 1. Schematic of a metal/insulator/antiferromagnet heterostructure. The thermal expansion of Al induced by an fs laser generates an ultrafast strain wave propagating through MgO to NiO. This strain wave excites a THz AFM spin wave. The spin-wave frequency is manipulated by the electric field applied between the two electrodes on the opposite sides of NiO. This electric field breaks the inversion symmetry along the $y$-axis direction and generates DMI that shifts the dispersion curve of the AFM exchange spin wave. The magnetic anisotropy constant, DMI vector, and the electric field are all indicated.

An optoacoustic transducer can excite continuous ultrafast magnetization dynamics without laser-induced thermal effect, converting laser thermal energy to mechanical energy to trigger continuous ultrafast strain waves, and thus magnetization precession [17, 18]. A typical device for this optical-acoustic excitation of magnetization dynamics is a metal/insulator/magnet heterostructure [19, 20]. An fs laser irradiates the metal layer, exciting ultrafast thermal expansion that generates a strain wave. The insulator transmits the strain wave but blocks the heat conduction. The magnetic moments start to process after the strain wave reaches the magnetic medium. Based on this consideration, the optical-acoustic excitation of magnetization switching and THz FM spin waves has been proposed [20–22]. In general, this spin wave consists of an exchange spin-wave component and a magnetoelastic one, and the intersections of the dispersion curves of these two spin-wave components determine the spin-wave frequency [20]. The exchange spin wave displays a parabolic dispersion curve in ferromagnets, while the magnetoelastic spin wave has a linear dispersion curve crossing the origin. Therefore, there are only 1–2 intersection points, limiting the frequency range for the application of spin waves.

Unlike an FM spin wave, an AFM exchange spin wave exhibits a hyperbolic dispersion curve that approaches linear at high wavenumbers. On the other hand, the dispersion curve for an AFM exchange spin wave can also be shifted by manipulating the Dzyaloshinskii–Moriya interaction (DMI) [23]. In this way, the dispersion curve of an AFM exchange spin wave may be overlapped with that of a magnetoelastic spin wave in a wide frequency range. We propose optical-acoustic excitation of a broadband THz AFM spin wave in a metal/dielectric/antiferromagnets heterostructure based on this motivation.

2. Model and method

We consider an Al (10 nm)/MgO (400 nm)/NiO (400 nm) heterostructure (figure 1). Here Al converts the heat of an fs laser (800 nm wavelength, 20 fs pulse width, and 3.1 mJ cm$^{-2}$ pulse energy) to an ultrafast strain wave. MgO assists the strain-wave transmission but prevents thermal spreading into NiO. Here NiO is an ideal medium for a strain-induced spin-wave emitter owing to its large magnetostrictive coefficient (140 ppm) [24]. Thanks to the insulating property, an electric field can be applied on NiO to break the structural symmetry along the $y$-axis direction, which generates a DMI vector along the $z$-axis direction [25–27] and shifts the dispersion curve of the spin wave propagating along the $x$-axis direction [23]. A voltage is applied across NiO through the electrodes at the two sides of NiO.

We formulated an atomistic spin model to calculate the magnetization dynamics of an AFM chain composed of two FM sublattices. The discrete energy of an AFM chain is expressed as [28]:

$$E_m = J_S \sum_i \langle \vec{S}^{(i)} \rangle \cdot \langle \vec{S}^{(i+1)} \rangle - K_a \sum_i (\langle \vec{S}^{(i)} \rangle \cdot \vec{e}_z)^2 - D_a \sum_i \vec{e}_z \cdot (\vec{S}^{(i)} \times \vec{S}^{(i+1)}) + \sum_i \left\{ \sum_s B_{1s} \langle S_s^{(i)} \rangle^2 \varepsilon_{w} + \sum_{s \neq p} B_{2sp} S_s^{(i)} S_p^{(i)} \varepsilon_{sp} \right\}. \quad (1)$$
Here \( \vec{S}^i \) is the normalized spin magnetic moment at site \( i \) [i.e., \( |\vec{S}^i| = 1 \)], and \( S^{i(p)}_\text{DMI}(s, p = x, y, z) \) is the \( s(p) \) component of \( \vec{S}^i \). The first to the fourth terms represent exchange energy, anisotropy energy, DMI energy, and magnetoelastoeic energy, respectively. The parameters \( J, K, D, B_1, \text{and } B_2 \) are AFM exchange integral, easy-axis anisotropy energy, DMI energy, the first- and second-order magnetoelastic energy, respectively. For simplicity, we considered a polycrystalline or amorphous medium with a uniaxial anisotropy [29, 30]. The expression for the magnetoelastic energy in equation (1) is suitable for either an FM medium or an AFM one with a cubic structure [16, 31–33]. For a polycrystalline or amorphous medium, the magnetoelastic energy in equation (1) should be approximated as \( \sum_i B_{1a}(S^{i(0)}_i)^2 \varepsilon_{xx} \) since \( B_{1a} = B_2a \) and \( \varepsilon_{xx} = 0 \) except for \( s = p = x \) [33, 34].

We numerically solved the atomistic Landau–Lifshitz–Gilbert equation [35]:

\[
\frac{\partial \vec{S}(t)}{\partial t} = -\gamma \vec{S}(t) \times \vec{H}_{\text{eff}}(t) + \alpha \vec{S}(t) \times \frac{\partial \vec{S}(t)}{\partial t},
\]

where \( \gamma \) is the gyromagnetic ratio. In equation (2), \( \alpha \) is the Gilbert damping parameter. \( \vec{H}_{\text{eff}}(t) \) is the effective magnetic field that is related to the magnetic potential energy by \( \vec{H}_{\text{eff}} = -\frac{1}{\mu_0} \frac{\partial E}{\partial \vec{B}} \) (where \( \mu_0 \) is the vacuum permeability, and \( m \) is the magnetic moment in a unit cell and written as \( m = M_s d^2 \) with \( M_s \) the saturation magnetization and \( d \) the lattice constant. The \( \vec{H}_{\text{eff}} \) consisting of the exchange field \( \vec{H}_{\text{ex}}(t) \), the anisotropy field \( \vec{H}_{\text{A}} = \frac{K}{\mu_0} \vec{S}(t) \), the DMI induced field \( \vec{H}_{\text{DMI}} \), and the magnetoelastic field \( \vec{H}_{\text{ME}} \) can be derived from equation (1) as

\[
\vec{H}_{\text{ex}} = -\frac{1}{\mu_0} \frac{\partial E}{\partial \vec{S}(t)} + \vec{H}_{\text{A}}, \quad \vec{H}_{\text{DMI}} = \frac{1}{\mu_0} D \vec{S}(t) \times \vec{e}_x, \quad \text{and } \vec{H}_{\text{ME}} = \frac{1}{\mu_0} B_{1a}(S^{i(0)}_i)^2 \varepsilon_{xx} \vec{c}_i \text{ with } B_{1a} = -1.5 \lambda_{l000}(\epsilon_{11} - \epsilon_{12})d^2 \quad (\lambda_{l000} \text{ is saturation magnetostrictive coefficient, and } \epsilon_{11} \text{ and } \epsilon_{12} \text{ are elastic stiffness coefficients}).
\]

In the effective field for the exchange coupling and DMI, we only consider the contribution from the nearest magnetic moments. We used the rule for a triple scalar product to derive the DMI effective field for the magnetic moment at site \( i \) as follows:

\[
\vec{E}^{i(0)} = D_d \vec{e}_z \cdot (\vec{S}(t) \times \vec{S}(i+1) - \vec{S}(i) \times \vec{S}(i-1)) = D_d \vec{e}_z \cdot \vec{S}(t) \times \vec{S}(i+1) - \vec{S}(i-1) = D_d \vec{e}_z \cdot \vec{S}(t) \times \vec{S}(i+1) - \vec{S}(i-1)
\]

\[
\vec{E}_{\text{DMI}} = \sum_i \vec{H}_{\text{DMI}} = \sum_i \vec{H}_{\text{DMI}} \text{, and } |E_{\text{DMI}}| \text{ since } D_d \text{ is the DMI induced field}.
\]

The strain of NiO originates from the laser-induced thermal expansion of Al and MgO. The dynamics of the thermal expansion is calculated by \( \frac{\partial^2 u_x}{\partial t^2} = c_{11} \frac{\partial^2 u_x}{\partial x^2} - 3B \frac{\partial^2 T_i}{\partial x^2} \)

\[
(3)
\]

where \( u_x \) is the displacement along the \( x \)-axis direction, and \( \varepsilon_{xx} \) is defined as: \( \varepsilon_{xx} = \frac{\partial u_x}{\partial x} \), \( \rho, \beta \) and \( B \) are the density, thermal expansion coefficient, and bulk modulus of Al and MgO, respectively. \( T_i \) is the lattice temperature determined by the interaction between the laser and Al. The incident photons excite hot electrons that interfere with phonons and transfer the energy to lattice. This process is described by a two-temperature model [37]:

\[
C_e \frac{\partial T_e}{\partial t} = \kappa_e \frac{\partial T_e}{\partial x} - G(T_e - T_l) + Q(x, t);
\]

\[
(4)
\]

\[
C_l \frac{\partial T_l}{\partial t} = \kappa_l \frac{\partial T_l}{\partial x} + G(T_e - T_l).
\]

\[
(5)
\]

Here \( C_e \) and \( C_l \) are the thermal capacities of electrons and lattice, respectively. \( C_e \approx \gamma_e T_e \) for temperature far below Fermi temperature. \( \kappa_e \) is the electronic thermal conductivity. As compared to \( \kappa_e \), the lattice thermal conductivity \( \kappa_l \) is negligible [38]. \( G \) is an electron–photon coupling constant. \( Q \) depicts the energy distribution of a Gaussian laser beam as:

\[
Q = 2 \sqrt{\frac{\ln 2}{\pi}} \frac{(1-t_p)}{t_p} I_0 \exp \left( -\frac{4t_p}{t_p} 2(1-t_p) \right) - \frac{4}{\pi} \ln\left( 2(1-t_p) \right)
\]

where \( R \) is reflectivity, and \( \delta \) is optical absorption depth, and \( t_p \) is the duration of the laser pulse, and \( I_0 \) is pulse energy (the energy density for a single pulse of the laser) [39, 40].

In calculating the thermal expansion of Al and MgO, we focused on the laser-induced deformation without considering the magnetostrictive stress of NiO [20]. While in calculating the magnetostrictive stress of NiO, the thermal expansion is neglected owing to the thermal blocking by MgO. Under this assumption, the dynamics of the displacement of NiO is expressed as:

\[
\rho \frac{\partial^2 u_x}{\partial t^2} = c_{11} \frac{\partial^2 u_x}{\partial x^2} - B_{1a} \frac{\partial (m_i^2)}{\partial x}
\]

\[
(6)
\]
fast Fourier transformation (FFT, the sampling interval is 1 × 10⁻⁵ m⁻¹).

Figure 2(a) depicts the spatiotemporal temperature of the heterostructure under an fs laser.

3. Results and discussion

The parameters for solving equations (2)–(6) are as follows: \( \gamma_c = 135 \text{ J K}^{-2} \text{ m}^{-3}, k_c = 238 \text{ W K}^{-1} \text{ m}^{-1}, \)
\( C_1 = 2.43 \times 10^6 \text{ J K}^{-1} \text{ m}^{-3}, R = 0.852, \delta = 16.9 \text{ nm} \) [41], \( G = 5.69 \times 10^{17} \text{ W K}^{-1} \text{ m}^{-3} \) [42], \( \beta_{AI} = 2.3 \times 10^{-5} \text{ K}^{-1} \) [43], \( \rho_{AI} = 2700 \text{ kg m}^{-3}, c_{Al}^{11} = 107 \text{ GPa}, c_{Al}^{12} = 61 \text{ GPa}, c_{Al}^{22} = 28 \text{ GPa} \) [44], \( \rho_{MgO} = 3580 \text{ kg m}^{-3}, \)
\( c_{MgO}^{11} = 297 \text{ GPa}, c_{MgO}^{12} = 96 \text{ GPa}, c_{MgO}^{22} = 156 \text{ GPa} \) [45], \( \beta_{MgO} = 3.11 \times 10^{-5} \text{ K}^{-1} \) [46], \( \rho_{NiO} = 6828 \text{ kg m}^{-3}, c_{NiO}^{11} = 342.7 \text{ GPa}, c_{NiO}^{12} = 141.3 \text{ GPa} \) [47], \( J_s = 4.7 \times 10^{-2} \text{ eV} \) [48], \( M_s = 3.51 \times 10^5 \text{ A m}^{-1} \) [48], \( d = 0.42 \text{ nm} \) [28], \( \lambda = 140 \text{ ppm} \) [24], \( \gamma = 1.76 \times 10^{11} \text{ rad s}^{-1} \text{ T}^{-1}, K_d = 2.2 \times 10^{-4} \text{ eV} \) [28], \( \alpha = 1 \times 10^{-3} \).

Equations (2)–(6) were numerically solved using a typical Runge–Kutta method with a time step of 0.1 fs. The Al (10 nm)/MgO (400 nm)/NiO (400 nm) heterostructure is discretized into cubic cells with a size of 0.42 nm for NiO and 1 nm for Al and MgO. We exploited a continuity boundary condition at the Al/MgO and MgO/NiO interfaces. A high damping coefficient and acoustic absorbing boundary condition were posed at the right edge of NiO to suppress the reflection of spin waves and strain waves. We assume a slight deviation of \( S_0 \) from the z-axis by about 3° as an initial condition.

3. Results and discussion

Figure 2(a) depicts the spatiotemporal temperature of the heterostructure under an fs laser. \( T_e \) is dominant over \( T_h \) and the rising of \( T_e \) is localized in Al and MgO (inset of figure 2(a)). This result means that MgO successfully blocks the thermal diffusion into NiO. The laser-induced thermal expansion triggers the strain-wave propagating from Al to NiO with negligible reflection and decaying (figures 2(b) and (c)). The Néel vector starts to process as soon as the strain wave arrives at the left edge of NiO at around 100 ps (figures 2(c) and (d)). The temporal variation of \( n_i \) at \( x = 746 \text{ nm} \) is converted into a spectrum using the fast Fourier transformation (FFT, the sampling interval is 1 × 10⁻¹⁶ s) (figure 2(d)). The spectrum illustrates the \( n_i \) oscillating in a broad frequency band ranging from 0.5 THz to 2 THz and at a single frequency for a very weak peak around 3 THz. This behavior is different from the single-frequency FM spin wave by optical-acoustic excitation [20–22].

To explain the broadband of spin wave, we deduced the dispersion curve by 2D FFT (figures 3(a)–(c)). The dispersion curve for the magnetoelastic spin wave fits \( f = v_i k/2\pi \) with \( v_i \), the longitudinal sound velocity of NiO (\( v_i = \sqrt{c_{11}/\rho} \)) [20]. By using the method in reference [50], we derived the theoretical dispersion for the exchange spin wave: \( f_{\pm} = \sqrt{\alpha_i^2 + (\pm D k + K_x)}/2\pi, \) where \( a = \frac{\gamma_{MgO} c_{MgO}^{11}\gamma_{NiO} c_{NiO}^{11}}{2\lambda}, \alpha_i = A - \frac{K_x}{\pi} \) with \( L = \frac{A}{2MgO}, D = \frac{4\gamma_i c_{NiO}^{11}}{\pi}, \) and \( K_x = \frac{4\alpha_i}{\pi}. \) Here \( f_{\pm} \) is the frequency for the right (left)-handed spin-wave mode. One can see that the simulated dispersion curve for an exchange spin wave satisfies the derived one at a low and moderate \( k \). At a high \( k \), the simulated dispersion deviates from the derived formula and the
frequency approaches a saturation value, since the wavelength approaches the limit of the cell size. This is consistent with the reported result of atomistic simulation [49].

Under zero DMI, the exchange spin wave has a hyperbolic-type dispersion curve with a gap at \( k = 0 \), while the magnetoelastic spin wave dispersion is a straight line crossing the origin. In this case, there is only one intersection for these two dispersion curves (figure 3(c)). Nevertheless, under a positive (negative) \( D \), the dispersion curve of the right (left)-handed mode shifts right and overlaps with that of magnetoelastic spin wave in a wide frequency range (from 0.5 THz to 2 THz) (figures 3(a) and (b)). While the dispersion curve of the left-handed (right-handed) mode still intersects with that of the exchange spin wave at a single frequency (3 THz). On the other hand, the increase of \( D \) also raises the frequency of the left-handed mode but lowers that of the right-handed one, enlarging the frequency gap between the two modes (figure 3(d)).

The original AFM spin waves are composed of opposite chirality components (figure 4(a)). These two spin-wave components can be separated using an analog low-pass and high-pass filter (inset of figure 4(a)). We simulated this mode filtering for the oscillation of \( \Delta n_x \) and \( \Delta n_y \) at \( x = 746 \) nm for \( D = 1.55 \) meV. (\( \Delta n_x(y) = n_x(y) - n_x(y)0 \) with \( n_x(y)0 \) the initial \( x(y) \) component of Néel vector for exhibiting the oscillation amplitude.) The low-frequency (\( f = 0.48 \) THz) oscillation of \( \Delta n_x \) and \( \Delta n_y \) and the (\( \Delta n_x, \Delta n_y \)) trajectory show right-handed precession (figures 4(b) and (c)). While the spin wave is passing, the high-pass filter exhibits left-handed precession with slower decaying (figures 4(d) and (e)). However, the strength for the left-handed precession is significantly weaker than that of the right-handed one by several magnitudes. This indicates that the AFM spin wave exhibits almost a single chirality tuned by an external electric field. This chiral spin wave can be exploited in information storage and computation.

Finally, we briefly discuss about the feasibility of the proposed design. We estimated the voltage for generating the DMI vector based on the formula [27]: \( \vec{D}_a = -J_d \frac{E_{SO}}{E_{SO}} \vec{E} \times \vec{e}_{12} \). Here \( J_d, e, d, E_{SO}, \vec{E}, \) and \( \vec{e}_{12} \) are the exchange integral (4.7 \( \times \) 10\(^{-2} \) eV), the charge of an electron (1.6 \( \times \) 10\(^{-19} \) C), the lattice constant (0.42 nm), the energy of spin–orbit coupling, the electric-field strength, and the unit vector for connecting two neighboring sites, respectively. The \( E_{SO} \) is given by \( E_{SO} = \frac{\hbar^2}{2m_e\lambda} \), where \( \hbar \) is the reduced Plank constant, \( m_e \) is the mass of the electron, and \( \lambda \) is the spin-orbital coupling constant. \( E_{SO} \) is typically on the order of 1 eV [25]. Based on the parameters and device dimension, we show that to generate a \( D_a \) of 1 \( \times \) 10\(^{-3} \) eV, the electric-field strength and the applied voltage are at the magnitude of 10\(^7 \) V m\(^{-1} \) and V, respectively. This is close to the estimation about the DMI energy of an insulating AFM medium by Wang et al [51] and Kim et al [52].
4. Conclusion

We propose broadband THz AFM spin-wave excitation based on optoacoustic conversion in Al/MgO/NiO heterostructure. The spin-wave frequency is determined by the intersection of the dispersion curve of an AFM exchange spin wave and that of a magnetoelastic one. By manipulating DMI, the dispersion curves of these two spin-wave components can overlap in a wide frequency range, and analog low-pass and high-pass filters can separate the two spin-wave components with opposite chirality. This work paves a way to develop an ultrafast AFM information device for storage and computation.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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