

Terahertz radiation from coherent antiferromagnetic magnons excited by femtosecond laser pulses

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We report on the observation of terahertz (THz) radiation emitted from antiferromagnetic (AFM) single-crystal nickel oxide irradiated with femtosecond laser pulses. Periodic oscillations observed in the THz waveforms are assigned to the radiation from coherent AFM magnons excited by the laser pulses. Impulsive stimulated Raman scattering process is a possible mechanism of the coherent AFM magnon excitation by the laser pulses. The excited magnons in NiO generate THz waves by magnetic dipole radiation, which is the inverse process of AFM resonance absorption of THz waves. © 2010 American Institute of Physics. [doi:10.1063/1.3436635]

The observation of terahertz (THz) radiation from materials excited by optical laser pulses, namely, “THz emission spectroscopy,” allows the ultrafast dynamics of the microscopic properties of materials to be investigated. This technique has previously been used to study the dynamics of photoexcited carriers in semiconductors¹ and superconductors,² as well as the band structure of semiconductor devices.¹ In addition, coherent phonons have been investigated by THz emission spectroscopy.^{3,4} In almost all previous research, surge currents and/or changes in polarizability originating from the motion of the charges triggered by the optical pulses have been investigated.

In recent years, sub-THz radiation emitted from ferromagnetic metals irradiated with femtosecond optical pulses, that is, magnetic dipole radiation of fast demagnetization induced by laser pulse irradiation has been reported.^{5,6} These previous findings indicate that THz emission spectroscopy can provide a means of studying the ultrafast response of electron spins directly. However, no other studies regarding the THz radiation from electron spins have been reported at present.

Meanwhile, the ultrafast control and detection of the electron spins in magnetic materials through the use of femtosecond optical pulses have been intensively investigated with regard to fast magnetic recording and fast spintronics.^{7–15} In particular, antiferromagnets are among the most promising materials for such devices because of their lack of macroscopic magnetization^{14,15} and higher magnetic resonance frequency.

In this paper, we report that THz pulses with periodic oscillations are radiated from antiferromagnetic (AFM) single-crystal NiO irradiated with linearly polarized femtosecond laser pulses. We conclude that the periodic oscillations are radiation from coherent AFM magnons excited by the laser pulses. Our results show that THz emission spectroscopy is a potentially useful tool for investigating ultrafast electron spin dynamics in various magnetic materials.

In this study, a regeneratively amplified Ti:sapphire laser with a repetition rate of 1 kHz, a pulse duration of about 120 fs and a center wavelength of 800 nm was used. The linearly polarized femtosecond laser pulses were focused on the sample at an incident angle of about 20° [Fig. 1(c)]. The

beam diameter on the sample was approximately 1 mm. The waveforms of THz radiation in the forward direction were measured by free-space electro-optic (EO) sampling using a 2.5 mm thick ZnTe(110) crystal.¹⁶

We used AFM NiO(110) single crystals as samples. Above the Neel temperature ($T_N \sim 523$ K), NiO has a rock-salt structure with inversion symmetry.¹⁷ Below T_N , the spins of Ni²⁺ ions along the $\langle 11-2 \rangle$ direction are ferromagnetically ordered within $\{111\}$ planes. Since ferromagnetic $\{111\}$ layers are reversed in adjacent $\{111\}$ planes, the crystal is AFM and thus characterized by a magnetic structure with easy-plane magnetic anisotropy. The spin order induces a small contraction of the cubic structure along the $\langle 111 \rangle$ direction, perpendicular to the ferromagnetic layers. Moreover, small distortions along the spin direction $\langle 11-2 \rangle$ also exist. Even if the contraction and distortions in the crystal are considered, the inversion symmetry is maintained. NiO crystals are usually twinned with four types of T-domain that are equivalent

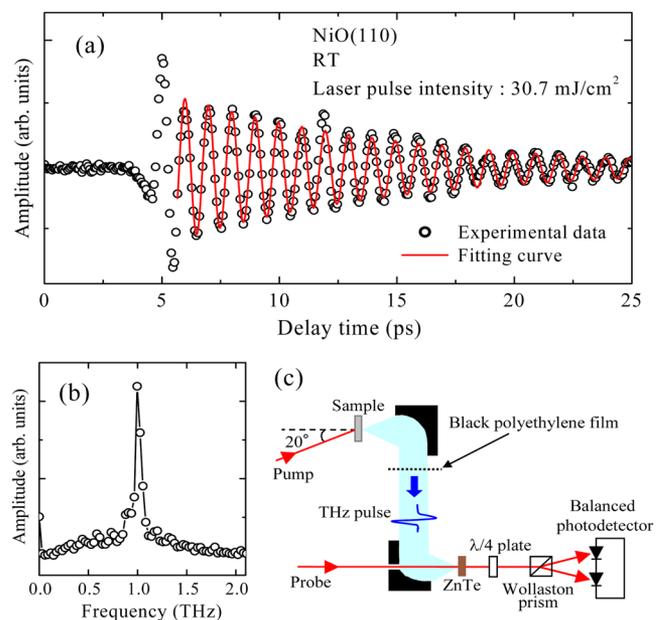


FIG. 1. (Color online) (a) Time-domain waveform of THz radiation from NiO(110). Solid curve is the fit obtained by using Eq. (3). (b) Frequency-domain spectrum of THz wave radiated from NiO(110). (c) Schematic diagram of experimental setup.

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to the {111} planes. For each T-domain, three types of S-domains with three possible orientations of spin can exist. Therefore, NiO crystals have 12 orientational domains.¹⁷ In all samples, domains with a typical size of $\sim 1 \mu\text{m}^2$ were observed by polarizing microscopy. The thickness of the samples used in the THz radiation experiments was $\sim 300 \mu\text{m}$. The absorption coefficient of NiO at 800 nm (1.55 eV) is $\sim 100 \text{ cm}^{-1}$ at 300 K.¹⁸ In all measurements in the present study, the intensity of the excitation laser pulse was well below the sample damage threshold of $\sim 60 \text{ mJ/cm}^2$.

Figure 1(a) shows a waveform of the THz pulse radiated from NiO(110) excited by linearly polarized femtosecond laser pulses with an intensity of 30.7 mJ/cm^2 at room temperature. The laser pulses excite the sample at a delay time of $\sim 5 \text{ ps}$. Oscillations with a period of $\sim 1 \text{ ps}$ ($\sim 1 \text{ THz}$) following a main pulse at a delay time of $\sim 5 \text{ ps}$ were observed. The dependence of the amplitude of the main pulse on the laser intensity and sample orientation (not shown) suggests that the generation of the main pulse can be attributed to a second-order nonlinear optical effect such as optical rectification induced at a surface with AFM ordering.¹⁹ The origin of the main pulse will be discussed elsewhere. In this paper, we focus on the oscillations ($\sim 1 \text{ THz}$ at RT) after the main pulse. The spectrum [Fig. 1(b)] obtained from the Fourier transform of the waveform clearly shows that the oscillations are the electromagnetic wave radiation with a frequency of $\sim 1 \text{ THz}$. The amplitude of the oscillations is almost proportional to the laser pulse intensity as shown in Fig. 3.

The solid circles in Fig. 2 show the temperature dependence of the frequency of the oscillations. The intensity of the excitation pulses was 21.5 mJ/cm^2 (5–280 K) and 30.7 mJ/cm^2 (300–400 K). The frequency of the oscillations normalized with respect to the antiferromagnetic resonance (AFMR) frequency, $\omega_{\text{AFMR}}(0)/2\pi = 1.07 \text{ THz}$ at 0 K obtained from transmission measurements in the THz frequency region mentioned later, is plotted as a function of normalized temperature T/T_N ($T_N = 523 \text{ K}$ is the Neel temperature of NiO). The AFMR frequency $\omega_{\text{AFMR}}(0)/2\pi = 1.07 \text{ THz}$ at 0 K obtained by extrapolation agrees well

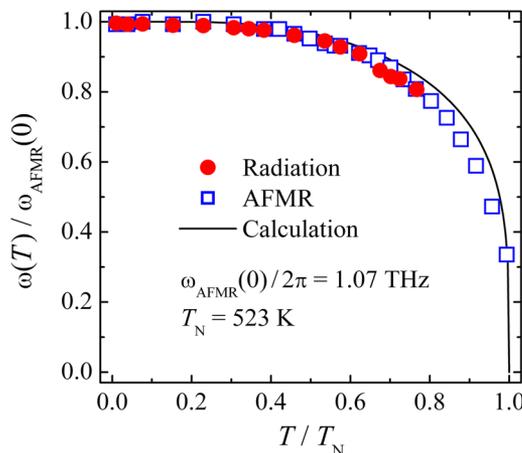


FIG. 2. (Color online) Temperature dependence of the frequency of the oscillations radiated from NiO (solid circles) compared with the temperature dependence of the AFMR frequency (open squares). Solid curve shows temperature dependence of the AFMR frequency calculated using a model based on the molecular field approximation.

with the previous result ($\sim 1.10 \text{ THz}$ at 2 K) reported in Ref. 21. For comparison, the temperature dependence of the AFMR frequency of NiO obtained from the transmission measurements by conventional time-domain spectroscopy in the THz frequency region is shown as the open squares in Fig. 2. The electromagnetic wave at the AFMR angular frequency ω_{AFMR} is absorbed, which leads to the excitation of AFM magnons with a wave number of almost zero via a magnetic dipole transition. In an AFM system composed of two sublattices with magnetization M , ω_{AFMR} is given by

$$\omega_{\text{AFMR}} = \gamma [H_A(2H_E + H_A)]^{1/2}, \quad (1)$$

where γ is the geomagnetic ratio, $H_A = K/M$ is the anisotropy field, $H_E = \lambda M$ is the exchange field, K is the anisotropy constant, and λ is the molecular field coefficient.^{20,21} The solid curve in Fig. 2 shows the temperature dependence of the AFMR frequency of NiO obtained using Eq. (1) within the molecular field approximation assuming that the temperature dependence of the sublattice magnetization is described by a modified Brillouin function with a total spin quantum number $S=1$. In total, the temperature dependence of the observed AFMR frequency (open squares) is well reproduced by the above-mentioned model. At $T/T_N > 0.7$, the observed frequencies are slightly lower than the predictions of Eq. (1), which is consistent with a previous study by Sievers and Tinkham,²¹ who noted that the temperature dependence of the sublattice magnetization of NiO deviates from a simple Brillouin function.

The origin of the oscillations following the main pulse in Fig. 1(a) is explained as follows. As shown in Fig. 2, the temperature dependence of the radiation frequency nearly coincides with that of the AFMR frequency. Therefore, coherent radiation with a frequency of $\sim 1 \text{ THz}$ at RT is emitted from coherent AFM magnons with a wave number k of ~ 0 , upon excitation by the femtosecond laser pulses. Since the magnetic moments of the two sublattices in NiO are not perfectly antiparallel,^{20,21} an effective magnetic moment $m(t)$ oscillating at the frequency of the magnons with $k \sim 0$ which is same as that of AFMR is produced when the magnons with $k \sim 0$ are excited. The mechanism of the excitation of the magnons by the optical pulses is discussed later. The coherent radiation following the main pulse, as shown in Fig. 1(a), is attributed to the radiation from the effective magnetic dipole moment $m(t)$ oscillating at the magnon frequency. In other words, the radiation mechanism is considered as the inverse process of AFMR absorption of THz waves. The electric field \vec{E} of the electromagnetic wave radiated from $m(t)$ via magnetic dipole radiation is given by

$$\vec{E} \approx \frac{-1}{c^2 R} \left[\frac{\partial^2 m(t)}{\partial t^2} \right] (\hat{n} \times \hat{R}), \quad (2)$$

where c is the velocity of light, R is the distance from the magnetic dipole, \hat{R} is the unit vector of the direction of R , and \hat{n} is the unit vector of the direction of magnetic dipole oscillation.^{6,22} $m(t)$ can be written as $m(t) = m_0 \sin(\omega_M t + \varphi) \exp(-t/\tau)$, where m_0 , τ , and φ are the amplitude, the relaxation time, and the phase of the magnon with angular frequency of ω_M , respectively. According to Eq. (2), the amplitude E_{THz} of the radiated THz pulses is given by

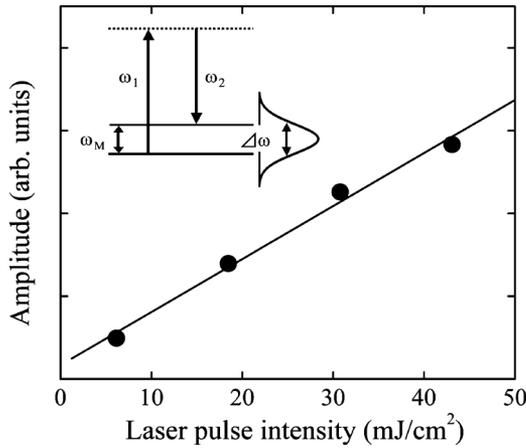


FIG. 3. Laser pulse intensity dependence of the amplitude of the radiation from coherent AFM magnons (solid circles). Solid line shows the result of the fit. Inset shows energy diagram describing coherent magnon excitation via ISRS.

$$E_{\text{THz}} \propto \omega_M^2 \sin(\omega_M t + \Phi) \exp\left(-\frac{t}{\tau}\right), \quad (3)$$

where Φ is the phase of the radiated electromagnetic wave. The waveform after a delay time of 5.6 ps in Fig. 1(a) is well fit by Eq. (3), yielding the parameters of $\tau=9.7$ ps and $\omega_M/2\pi=1.0$ THz. The result of the fit is shown as the solid curve in Fig. 1(a).

Next, we will discuss the mechanism of the coherent AFM magnon excitation by the femtosecond laser pulses. As shown in Fig. 3, the amplitude of the radiation from the AFM magnons is almost linearly related to the intensity of the excitation optical pulses. This suggests that a possible mechanism of the coherent AFM magnon excitation is an impulsive stimulated Raman scattering (ISRS) process²³ via virtual states with spin-orbit coupling. The energy diagram of the excitation process is presented in the inset of Fig. 3. Photons with energies of both $\hbar\omega_1$ and $\hbar\omega_2=\hbar\omega_1-\hbar\omega_M$ are contained in the excitation laser pulses, thus allowing for induction of the ISRS process. Owing to the virtual states with spin-orbit coupling, a spin-flip process becomes possible,²⁴ which leads to the excitation of the magnons.

It should be noted that the excitation of the AFM magnons via the ISRS process can be viewed as inverse Faraday^{8–11} and inverse Cotton–Mouton¹² effects when circularly and linearly polarized excitation pulses are used, respectively. Since linearly polarized pulses are used in this work, the inverse Cotton–Mouton effect is considered to be dominant. However, we are unable to strictly distinguish between these two effects because of the existence of linear magnetic birefringence of NiO in the AFM state.²⁵ A part of the incident excitation pulses is thought to be converted to

circularly polarized light through magnetic birefringence, and thus the two effects can be mixed in the excitation of the AFM magnons in these experiments.

In conclusion, we have carried out THz emission spectroscopy on AFM NiO single crystals. The observed periodic oscillations in the waveforms are attributed to magnetic dipole radiation from coherent AFM magnons excited by the femtosecond laser pulses. The radiation can be viewed as the inverse process of AFMR absorption. The mechanism of the coherent AFM magnon excitation can be attributed to the ISRS process induced by the femtosecond laser pulses. THz emission spectroscopy on AFM materials is a useful technique for studying the fast response of their electron spins.

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