Coherent Control of Spin Precession Motion with Impulsive Magnetic Fields of Half-Cycle Terahertz Radiation

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Coherent control of the precession motion of magnetizations in a single crystal $YFeO_3$ with double half-cycle pulse terahertz waves was demonstrated. Quasiferromagnetic (0.299 THz) and quasiantiferromagnetic (0.527 THz) precession modes were selectively excited by choosing an appropriate interval of two pulses and were observed as free induction decay (FID) signals from the spin system. By observing the circularly polarized FID signals due to ferromagnetic resonance, we also succeeded in confirming directly the energy storage in the spin system and recovery from that to the electromagnetic radiation.

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Motivated by the rapid progress in the femtosecond laser spectroscopy, a number of studies on coherent control of quantum mechanical oscillations have been made. Real time observations of dynamical behavior under coherent control have been applied to various targets including nuclear wave packets in molecules, electronic wave packets in atoms [1], and elementary excitations in condensed matter such as phonons [2–4], excitons [5], and magnons [6–11]. In addition to a deeper comprehension of the wave nature of matter and the attainment of an otherwise impossible state of matter, coherent control is expected to play an important role in applicative fields such as information processing and storage mediated by quantum systems [1]. For example, manipulation of spins in ultrafast time scales is one of the vital techniques required for advancements in optical memory, quantum computing, and spintronics technologies [12], making coherent control of spin systems an intriguing subject. Preceding studies on spin precession excitations with ultrashort laser pulses were based mainly on indirect excitations of spin systems that take place as a consequence of dielectric interaction with photons such as spin polarized carrier generations [6], change in distribution of the photoexcited holes [7,13], optical Stark effect [8], inverse Faraday (or Cotton-Mouton) effect which is interpreted as an exchange of angular momentum between lights and spins through Raman process [9,11,14,15], and excitations of *d-d* transition [10] as well as thermal effect [15]. However, reports on the direct excitation of spin precession motion by means of magnetic field components in light waves are limited [16], and coherent control in this scheme has been hitherto unreported to the best knowledge of the authors.

The most commonly observed coherent control is that in phonon system, so-called coherent phonon experiment. The phonon oscillation coherently induced by the first pulse can be either completely damped or enhanced to twice the initial amplitude by the second pulse by adjusting the time interval of two pulses [2–4]. Coherent control of spin precessions can be performed in this fashion as well [6–11]. In these kinds of experiments, there is a still remaining discussion on whether they are really coherent manipulation of the oscillator itself rather than superposition of two separately excited states with inverted phases [9]. Assuming that such examples are truly coherent control of elementary excitations, in the destructive case, i.e., when the second pump pulse attenuates the excitation caused by the first pulse, energy given from the first pulse to the phonon system is expected to be transferred back to the second pumping light. However, up until now, the energy conservation between photons and elementary excitations has not been proven experimentally. This is because in the preceding experiments with visible light, the energy incident on the sample is many orders of magnitude larger than that stored in the elementary excitation system. Also, even if the input and output energies are precisely monitored, observation of energy conservation is not guaranteed because additional energy dissipation channels such as carrier generation and heating may exist in such cases. Since precession motions of the spins in our study are excited and monitored through the terahertz (THz) waves coupled directly with the magnetic moments in the sample, insight into the conservation of energy is expected to be obtained.

Here, by applying the method of THz time domain spectroscopy (TDS), we demonstrate coherent control of spin precession motion in a magnetically ordered material with excitations caused by a sequence of two pulses of THz waves. As a THz wave enters a magnetically ordered material, the magnetic field component of the THz wave acts as a subpicosecond magnetic pulse. As shown in Fig. 1(a), when a half-cycle magnetic field acts on such material, the magnetic moment experiences a force which equals to the cross product of the magnetic moment and the impulsive field, and is tilted. After this tipping, the magnetic moment starts to precess around the effective magnetic field in the z axis direction [Fig. 1(a)]. The induced precessions of the spins are expected to radiate free induction decay (FID) signal as a circularly polarized electromagnetic wave with a frequency

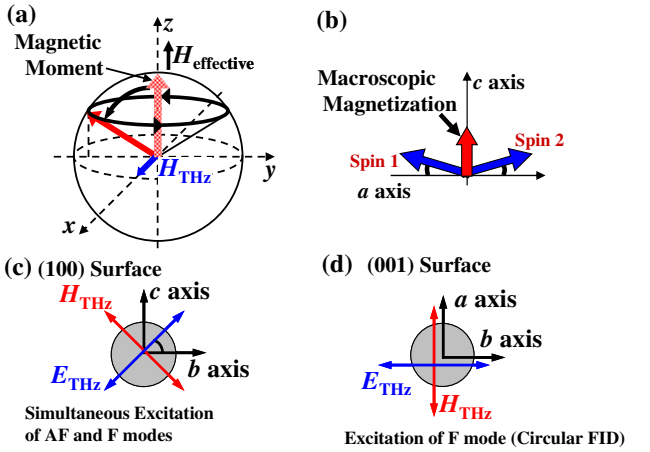

FIG. 1 (color online). (a) Illustration showing the dynamics of magnetic moment. The magnetic moment is tilted within y-z plane by the impulsive magnetic field along x axis (blue arrow), and begins precession motion around z axis. (b) Ordering of Fe^{3+} magnetic moments in $YFeO_3$. Illustrations (c) and (d) show the crystal axes and directions of electric and magnetic fields of the THz wave in the experiments of Figs. 2 and 3, respectively.

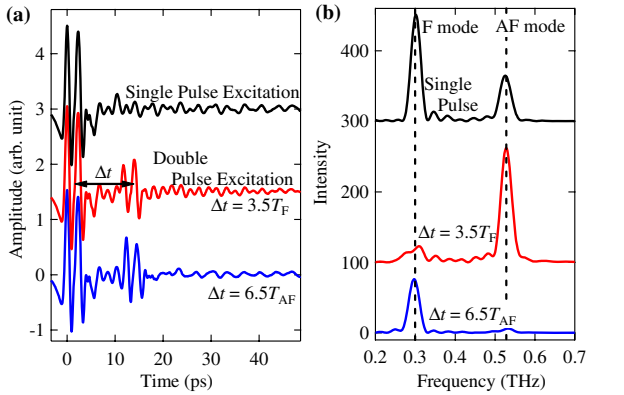

FIG. 2 (color online). (a) Temporal waveforms of transmitted THz wave through a-cut plane parallel plate of $YFeO_3$. (b) Spectra obtained by Fourier transformation from 18 to 48 ps in Fig. 2(a).

equal to the precession frequency, which persists within the transverse relaxation time.

The samples used in this study are single crystals of yttrium orthoferrite $YFeO_3$ grown by the floating zone method (Crystal Systems Corporation). $YFeO_3$ is an antiferromagnet below the Néel temperature $T_N = 645$ K [17], and nearest neighbors of Fe^{3+} ions are ordered in antiparallel alignment along the a axis of the orthorhombic crystal lattice [18]. However, as Fig. 1(b) shows, $YFeO_3$ possesses macroscopic magnetization in the direction of the c axis ascribed to canting of Fe^{3+} ion magnetic moments caused by antisymmetric exchange interaction [18]. $YFeO_3$ is known to have two branches of antiferromagnetic resonance in sub-THz frequency range. One is called quasiantiferromagnetic mode (AF mode) and has resonant frequency of 0.527 THz at 300 K. The other branch with its resonant frequency at 0.299 THz (300 K) is called quasiferromagnetic mode (F mode), which is interpreted as a precession motion of the macroscopic magnetization [19]. Distinct from the ordinary antiferromagnetic resonance, owing to this macroscopic magnetization, we are able to observe the irradiation of circularly polarized FID in the F mode. In this study, single crystal $YFeO_3$ platelets with their plane normal to the a axis and c axis were prepared. Both samples were fixed to a metal aperture with a diameter of 4 mm, and the thickness of a-cut and c-cut plane parallel plates are 1.7 and 1.4 mm, respectively.

Transmission type THz TDS measurements with these samples were conducted at room temperature. Emission and detection of the THz waves were done with dipole type low temperature grown GaAs photoconductive antennas that emit and detect THz waves with horizontally polarized electric field component. These antennas were excited with mode-locked Ti:sapphire ultrashort laser pulses (12 fs) with their center wavelength at 800 nm and the repetition rate at 75 MHz. As it will be seen in the following part of this report, the emitted THz wave is nearly single sided in polarity, and is regarded as a half-cycle pulse when it acts on the spin system. THz signals in the polarizations parallel and perpendicular to the antennas were evaluated from the sum and difference of the signals in $\pm 45°$ polarization angles, respectively [16]. The first and the second pulses of THz wave were separated by spatially splitting the pumping laser beam in its cross section with a pair of hemicircular shaped mirrors. The pair of mirrors was put with their edge close together and the laser beam was reflected in the proximity of these edges so each mirror reflects some portion of the incoming laser beam. The time delay Δt between the two pulses was adjusted by moving one of the two mirrors back and forth. By changing the cross sectional area of the laser reflected by each mirror, amplitude ratio between the two pulses was adjusted.

Figure 2(a) shows the transmitted temporal waveforms obtained by measurements with the a axis normal plane sample. The b axis of the sample was set at 45° from the electric vector of the incident THz wave [Fig. 1(c)]. In this configuration, oscillation with both F mode and AF mode resonant frequency can be observed simultaneously. As seen in Fig. 2(a), the main peak of the THz pulse is followed by oscillations that result from FID of magnetic resonances in the sample. The main peaks at 0 ps in Fig. 2(a) are split because the arrival time of the transmitted THz waves in $E||b$ and $E||c$ polarizations differs due to the birefringence in the sample ($n_b = 4.57$ and $n_c = 5.05$) [17]. This also applies for the second pulses at around 13 ps. From Fig. 2(b) which shows the Fourier transformed spectra of the oscillation components in Fig. 2(a), two peaks at around 0.3 and 0.53 THz can be observed when the excitation occurs only once. Thus, it was confirmed that the single pulse excitation in this configuration actually results in excitation of both F mode and AF mode magnetic resonance. The waveforms in Fig. 2(a) labeled as

$\Delta t = 3.5T_F (= 6.08T_{AF})$ and $\Delta t = 6.5T_{AF} (= 3.74T_F)$ were obtained by double-pulse excitation with two pulses temporally separated by time delay Δt, where $T_F = 3.3$ ps and $T_{AF} = 1.9$ ps are oscillation period of F mode and AF mode resonance, respectively. In Fig. 2(a), by comparing the temporal waveforms of single pulse excitation, $\Delta t = 3.5T_F$, and $\Delta t = 6.5T_{AF}$, obvious changes in the behavior of the oscillations are recognized. As Fig. 2(b) shows, F mode resonance is canceled and only the AF mode resonance remains when the second excitation takes place at $\Delta t = 3.5T_F$. On the other hand, when the second pulse is delayed $\Delta t = 6.5T_{AF}$ from the first pulse, the AF mode disappears while the F mode resonance persists. Here, we tuned the amplitude of the two pulses so that one of the two modes would be completely canceled in the above cases.

With the c-cut plane parallel sample where the AF mode does not appear, coherent control of the F mode resonance was conducted. Again, the THz wave was linearly polarized, and this time the $YFeO_3$ sample was set so that electric and magnetic field components of the incident THz waves are parallel to the b axis and a axis of the crystal lattice, respectively [Fig. 1(d)]. Hereafter, we will refer to this polarization as $E||b$ polarization. During the measurements in this configuration, a static magnetic field of approximately 0.1 T was applied along the c axis to remove the quasiferromagnetic domain structure. Since the spontaneous magnetizations precess around the c axis direction in the F mode resonance, circularly polarized FID signal with an angular momentum parallel to c axis is expected to be emitted at F mode frequency. In fact, the temporal waveforms in Figs. 3(a) and 3(b) show oscillations with a frequency of approximately 0.3 THz and decay time of about 40 ps in both $E||b$ (parallel to the incident THz wave) and $E||a$ (perpendicular to the incident THz wave) polarizations, and there is a $\pi/2$ phase difference between oscillations in these polarizations, which indicates the emission of circularly polarized wave. Keep in mind that since the effective optical path lengths for $E||b$ and $E||a$ polarizations are not identical due to birefringence in the single crystals of $YFeO_3$ ($n_a = 4.80$, $n_b = 4.57$) [17], relation of the phase difference between the two polarizations depends on the thickness of the sample. By assuming that the length the circularly polarized light passes through the medium after its emission is, on the average, 0.7 mm, we estimated that the wave in $E||a$ polarization spends extra 0.5 ps to transmit through the sample when compared with the wave in $E||b$ polarization, and the birefringent effect was compensated in Figs. 3(a) and 3(b) by giving offset of -0.5 ps to the temporal waveforms of $E||a$ polarization.

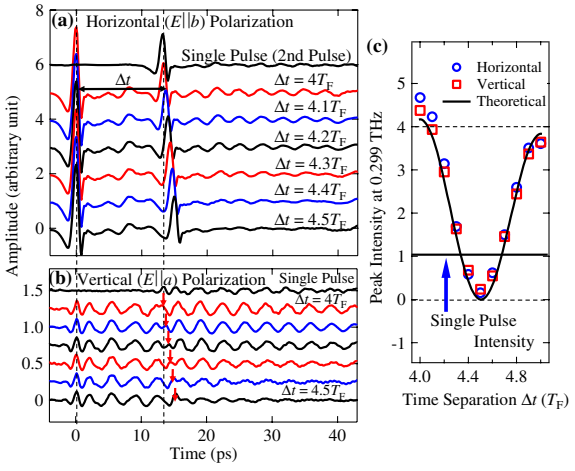

FIG. 3 (color online). Temporal waveforms of transmitted THz wave through c-cut plane parallel plate of $YFeO_3$ in (a) $E||b$, and (b) $E||a$ polarizations. (c) Normalized intensity at 0.299 THz peak obtained by Fourier transformation of the temporal waveforms from 18 to 43 ps.

As shown in Fig. 3, changing the separation time Δt enabled us to manipulate the amplitude and the phase of the oscillation in both $E||a$ and $E||b$ polarizations. When $\Delta t = 4.5T_F$, the emissions in both polarizations are annihilated. Similar to the previously described selective excitation experiment, the amplitude ratio of the two pulses was optimized so that the FID signal disappears in $\Delta t = 4.5T_F$. As Δt deviates from $\Delta t = 4.5T_F$, the amplitude of the emission gradually increases. Especially, when Δt is integer multiple of the oscillation period, the emission amplitude is doubly enhanced (quadruply in intensity) by the second excitation in comparison to the emission in single pulse excitation. These behaviors are also observed in the peak intensity of the oscillation at 0.299 THz normalized with that of the single pulse excitation shown in Fig. 3(c), which is obtained by Fourier transformation of Figs. 3(a) and 3(b). We also confirmed that the peak intensity in Fig. 3(c) fits well with a calculation which takes damping of the emission with 40 ps time constant into account.

In the excitation of precession motion, the energy of the impulsive magnetic field is transferred to the spin system instantaneously, resulting in amplitude diminishment of the incident electromagnetic wave. In analogy, when the second pulse stops the precession motion, it should receive energy from the spin system. This should be observed as an increase in the peak height of the magnetic field component in the second pulse. Actually as shown in Fig. 4(a), the peak height of the electric field component of the second pulse, which is considered to be proportional to the magnetic field component, is higher in the $\Delta t = 4.5T_F$ case than that of the single pulse irradiation (second pulse without a prepulse). We have also confirmed that the change in total energy of the main peak is comparable with the difference in the total energy of the FID radiation after the second excitation. If the second pulse had excited another set of spins and the disappearance of radiation were simply a cancellation of two out-of-phase radiation fields with one another, the energy of the second pulse should not differ from the single excitation case. Thus, this

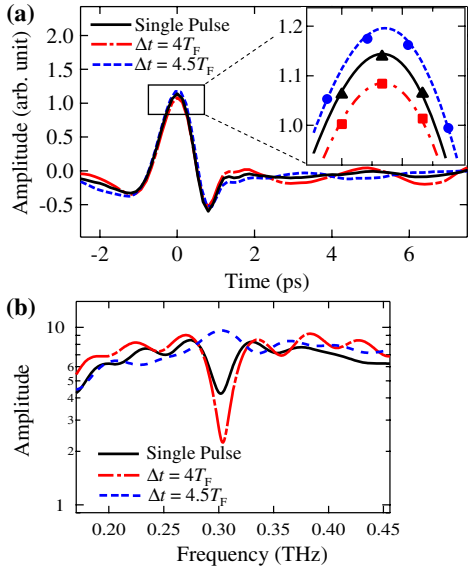

FIG. 4 (color online). (a) Main peak of the second pulses in $E||b$ polarization under single pulse excitation (second pulse without a first pulse), $\Delta t = 4T_F$, and $\Delta t = 4.5T_F$ condition. The inset shows magnified curves near the peak. (b) Fourier spectra of the three temporal waveforms shown in Figs. 3(a) and 4(a) within a time window with a width of 27 ps, starting from 2.5 ps before the main peak.

fact shows that the spin precession itself is really coherently controlled by THz waves.

In contrast, the peak height in $\Delta t = 4T_F$ case is lower in Fig. 4(a), showing that a larger amount of energy is transferred from the electromagnetic wave to the spin system than in the single pulse excitation case. These results show that the energy transfer takes place on each event of interaction of the pulsed magnetic field with the spin system. The existence of energy stored in the spin system is monitored as FID signals following the second pulse in $E||b$ polarization, where we can clearly see the FID without an overlapping of incident THz wave. In the case of $\Delta t = 4T_F$, amplitude is increased twice; i.e., the stored energy is 4 times larger, as shown in Fig. 3(c). This energy is released slowly as FID radiation after the second pulse and via other relaxation processes within the condensed matter.

Shown in Fig. 4(b) are the Fourier spectra of the second pulse. The spectrum of a second pulse without the first excitation have a dip at the resonant frequency of the F mode (0.299 THz) which corresponds to the FID signal in the temporal waveform following the main peak of the THz pulse in Fig. 3. Such emission of FID can also be considered as absorption of energy from THz wave, and in this perspective, the dip corresponds to the energy of electromagnetic wave absorbed by the spin system. The dip at 0.299 THz disappears when the temporal separation of the two pulses is $\Delta t = 4.5T_F$, which reflects the extinction of the FID signal after the second excitation. Such disappearance of the dip indicates that the energy brought to the spin system by the first pulse is instantaneously restored to the second pulse.

As a summary, we demonstrated coherent control of precession motion of magnetizations in a single crystal of $YFeO_3$ with THz double-pulse excitations. Precessions in both F mode and AF mode resonances were selectively excited by this method and especially in the coherent control of F mode resonance, manipulations of the collective motion of the spins were observed through circularly polarized FID signal from the spin system. Owing to the absence of the incident electromagnetic field in $E||a$ polarization, we were able to clearly observe the FID signal in the vicinity of the time origin. In addition, we verified the existence of the energy transfer process between the photon and the spin system which has been considered to take place during coherent control of quantum mechanical systems in condensed materials, utilizing simultaneous observation of the incident and FID fields in $E||b$ polarization. The combination of the technique shown here with high intensity THz pulses may result in much drastic control of spin systems such as magnetization reversal and spin reorientation.

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