Beating of Terahertz Pulse Induced Spin Precession in $ErFeO_3$

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Abstract: Terahertz pulse induced spin precession in $ErFeO_3$ was observed via the Faraday rotation of the visible probe pulse. Unreported splitting of the magnetic resonance was discovered and mechanism explaining this splitting is proposed.
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1. Introduction

Recent developments on the terahertz (THz) wave technology triggered intensive spectroscopic research on various materials in this frequency region. One of the newly developing topics is the ultrafast excitation of spins with the THz pulse [1-4], where the THz magnetic field component is used for instantaneously tilting the spins. Here, we report the long-lived spin precession with a beating induced by THz pulse in an orthoferrite $ErFeO_3$, which, to the best of our knowledge, has never been reported in the past.

Orthoferrites have four iron sublattices that order antiferromagnetically below Neel temperature of about 600 K. Owing to the antisymmetric exchange (Dzyaloshinskii-Moriya interaction), the angle between the nearest neighbor Fe^{3+} spins deviates slightly from 180 degree. This deviation gives rise to a weak macroscopic magnetization. In the temperature region studied here (10 K to 80 K), the easy axis for each sublattice spins is along the *c* axis and the macroscopic magnetization is along the *a* axis. There are two optically active magnetic resonance modes in the sub-THz region, namely quasiferromagnetic (F-) mode and quasiantiferromagnetic (AF-) mode. F mode resonance is interpreted as precession of the macroscopic magnetization, while AF mode can be seen as fluctuation in magnitude of the magnetization. F mode and AF mode is known to be excited with THz magnetic field perpendicular or parallel to the magnetization, respectively.

2. Result

The spin precession in $ErFeO_3$ single crystal sample (100 μm thick, (001) surface sample) induced by THz pulse was observed through time dependent Faraday rotation of the transmitting visible probe pulse. The Faraday rotation reflects the magnetization dynamics in the thickness direction of the sample, that is, the *c* axis direction. The THz pulse used for the spin excitation was generated from $LiNbO_3$ crystal with tilted pulse front technique.

Figure 1(a) shows the time dependent THz induced Faraday rotation obtained with this sample at 70 K. The polarization of the incident THz pulse was $H_{THz} \parallel b$ axis, which excites the F mode precession. As the figure shows, the spin precession is confirmed. However, unlike in the previous reports [1,2], the precession lasts for a long time and a 50 ps period beating of the oscillation is also observed. The Fourier spectrum of the oscillation in Fig. 1(b) reveals that the obtained signal consists of two separate sharp spectral peaks. The frequencies of the two peaks are

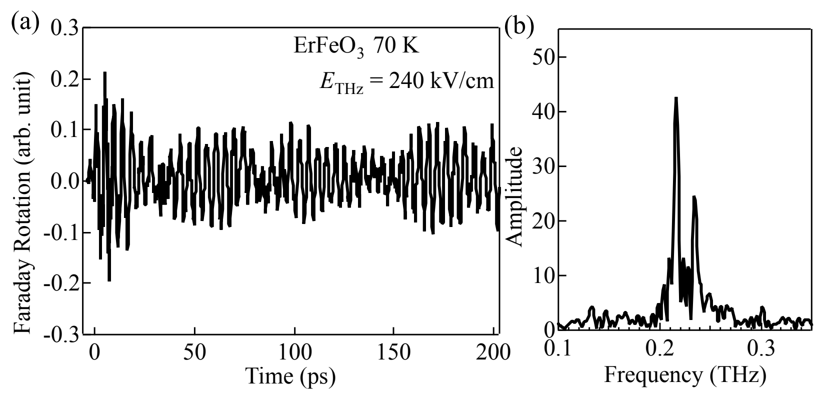

Fig. 1. (a) Time dependent THz induced Faraday rotation signal obtained with (001) $ErFeO_3$ single crystal sample at 70 K. (b) Fourier spectrum of the oscillation shown in (a).

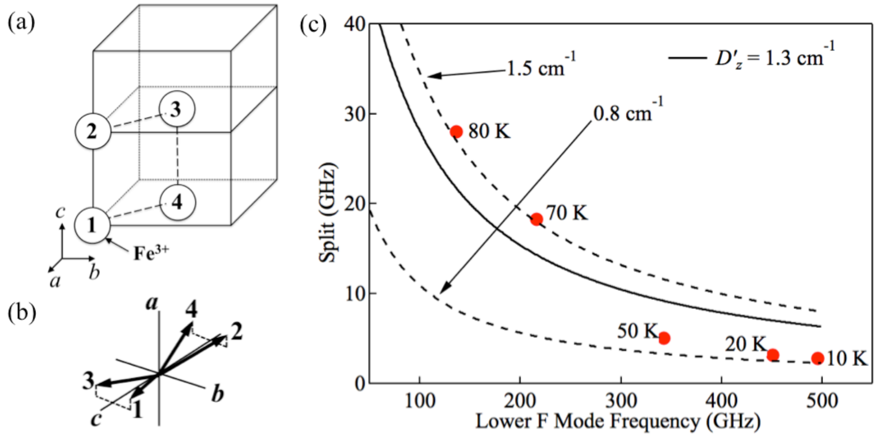

Fig. 2. (a) Crystallographic structure of $ErFeO_3$ and the position of the four Fe^{3+} sublattices. (b) The Fe^{3+} spin configuration in the temperature region below 90 K. (c) The splitting of the F mode resonance observed at each temperature. The lines show the calculated value for various D_z'.

approximately 0.216 THz and 0.234 THz (close to the F mode resonant frequency at 70 K reported previously [2]). The magnitude of this splitting corresponds to the beating period of 50 ps. However, as far as we are concerned, such splitting of the F mode resonance peak has never been reported in the past. This discrepancy with the previous researches may be due to the difference in the probed sample area. In the previous examples, comparatively large area (around 1 mm in diameter) of the sample was observed, and such fine structures of the spectrum may have been averaged out by spatial inhomogeneity. In the case of the present experiment, the probe spot diameter is roughly 30 μm, and such inhomogeneous effects may be suppressed.

Although there are several known mechanisms (standing spin wave or splitting due to the domain walls) that can cause the splitting of the magnetic resonance mode, the magnitude of the splitting with these mechanisms do not match with the observed splitting. Here, instead of these known mechanisms, a novel splitting mechanism is introduced. Considering the symmetry of the crystal, it is noticed that the Dzyaloshinskii-Moriya interaction works differently for different pairs of nearest neighbor spins among four spin sublattices [5, 6]. For example, when focusing on the spin 1 in Figs. 2(a) and (b), the nearest neighbor spins 2 and 4 interact with spin 1 differently. Normally, such differences are ignored for deriving the resonant frequency [7]. By taking this difference into consideration, the resonant frequencies were calculated as a function of lower branch F mode frequency, which depends on temperature. It was confirmed that in the spin configuration in Fig. 2(b), the z component of the Dzyaloshinskii vector D_z' working on spin pair 1-4 (and 2-3) causes the resonant frequency to split. Figure 2(c) shows the calculated value of the splitting along with the splitting obtained experimentally. The figure shows that the observed splitting can be explained reasonably with the proposed splitting mechanism using value of D_z' deduced from ref [7].

3. Conclusion

To summarize, THz pulse induced spin precession was observed via visible Faraday rotation in $ErFeO_3$ and extremely long-lived precession with an unknown splitting of the F mode resonant frequency was observed. We proposed a splitting mechanism based on the difference of the Dzyaloshinskii-Moriya interaction between different spin pairs, and confirmed that this theoretical model explains the observed phenomena reasonably.

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