

# Ultrafast Spin Dynamics in an Antiferromagnet NiO Observed in Pump-Probe and Terahertz Experiments

T. Moriyasu, S. Wakabayashi, and T. Kohmoto  
Graduate School of Science, Kobe University, Kobe 657-8501, Japan  
moriyaspin@gmail.com

**Abstract:** We observed the ultrafast spin dynamics in an antiferromagnet NiO. The dynamics of the antiferromagnetic magnons and the magnetostriction was studied using the pump-probe technique and THz-TDS.

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## 1. Introduction

The ultrafast spin dynamics and optical spin control in magnetic materials [1-4] are attractive topics because of the potential applications in the developments of ultrafast spin control, spintronics, quantum computing, and optical control of correlated spin systems. For the ultrafast spin control and device applications, solid-state materials are more desirable.

The magnon is one of elementary excitations in solid-state materials and plays important roles in magnetic materials. Usually, antiferromagnetic resonance frequencies, which are the precession frequencies of magnons, are higher than ferromagnetic resonance frequencies. Therefore, it is possible that antiferromagnets are used to realize the ultrafast spin control. In many cases, the antiferromagnetic resonance frequency lies in the terahertz region, and the spectroscopic investigation of antiferromagnets in this frequency region is very important.

In this study, we have investigated the magnons and magnetostriction in an antiferromagnetic 3d transition metal monoxide NiO using the pump-probe technique [5] and terahertz time-domain spectroscopy (THz-TDS).

## 2. Sample and experiment

NiO is a paramagnetic insulator with a cubic rock-salt structure above the Néel temperature 523 K. In the antiferromagnetic phase below the Néel temperature, the magnetic moments on Ni atoms align ferromagnetically within a (111) plane, and (111) planes are stacked antiferromagnetically in the direction normal to the (111) plane. The sample, whose thickness is 0.1 mm for pump-probe technique and 5 mm for THz-TDS, is placed in a temperature-controlled cryostat.

In the pump-probe experiment, the pump pulse (810 nm) is provided by a Ti:sapphire regenerative amplifier and the probe pulse (900 nm) by an optical parametric amplifier. The magnetization or the effective magnetic field induced by an inverse Faraday effect are instantaneously generated by the circularly polarized pump pulse. The generated magnetization is detected by a polarimeter as the change in the polarization of the linearly polarized probe pulse.

In the THz-TDS experiment, THz generation and detection are carried out via the use of a large-area electro-optic (EO) THz emitter and a standard EO detector, respectively. The optical pulses for THz generation and detection are provided by a Ti:sapphire regenerative amplifier. Their wavelength, pulse width, pulse energy, and repetition rate are 810 nm, 0.15 ps, 700  $\mu$ J, and 1 kHz, respectively.

## 3. Results and discussion

The typical magnon signals in NiO observed by the pump-probe technique [5] and THz-TDS are shown in Figs. 1(a) and 1(c), and their Fourier transform are shown in Figs. 1(b) and 1(d).

The observed temperature dependence of the antiferromagnetic magnon modes up to the Néel temperature in NiO is shown in Fig. 2. These modes are observed in a good accuracy by using the pump-probe and THz-TDS experiment than the result of Raman scattering and Brillouin scattering [6].

The magnetostrictive contribution to the refractive index was observed by a novel measuring method using THz-TDS. The change  $\Delta n$  in refractive index was obtained from the peak shift of the transmitted THz electric field, and its temperature dependence is shown in Fig. 3. The gray curve indicates the contribution from the thermal expansion calculated from the internal energy of phonons. The deviation from the calculated curve observed below the Néel temperature is considered to be a magnetostrictive contribution.

Our experimental results show that the time-domain spectroscopy has a large potential for the sensitive and accurate measurements in magnetic materials.

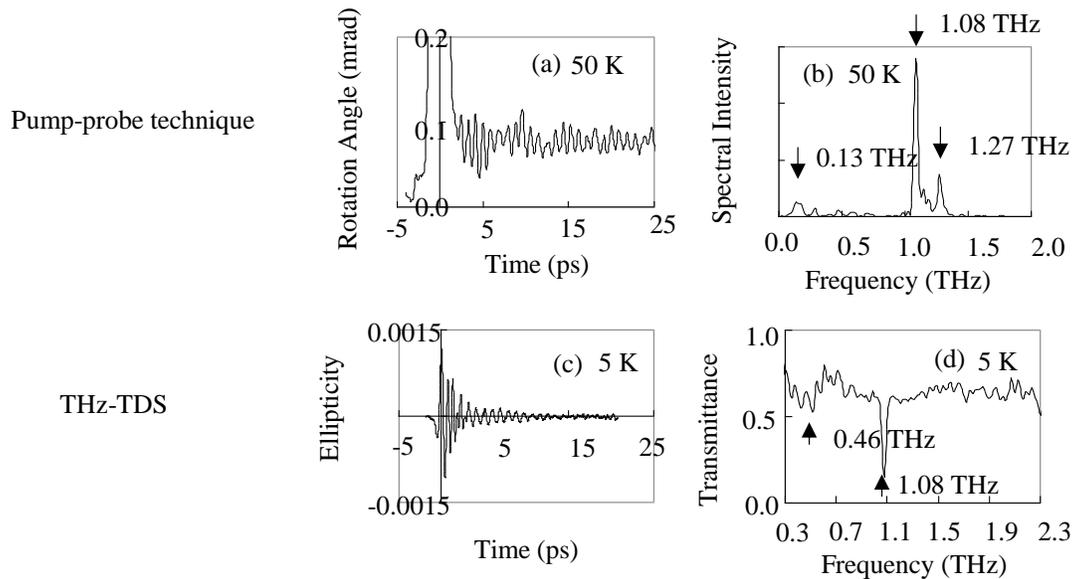


Fig. 1. Magnon signals in NiO observed by (a) the pump-probe technique and (c) THz-TDS. (b) and (d) are the Fourier transform of the signals in (a) and (c).

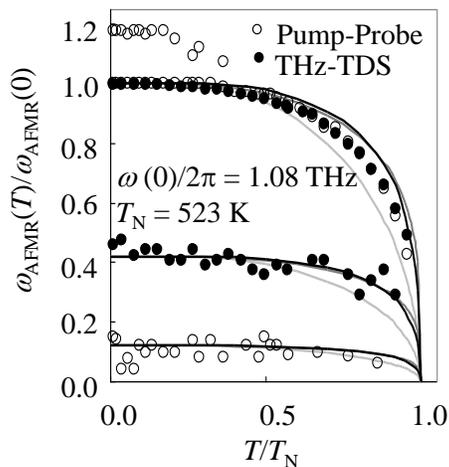


Fig. 2. Temperature dependence of the magnon frequencies observed in NiO.

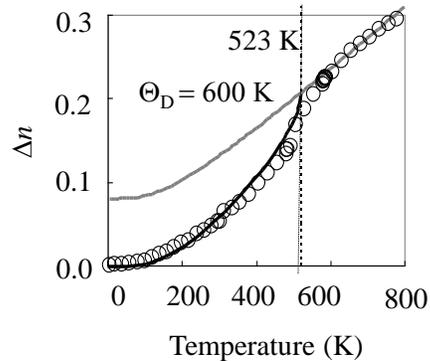


Fig. 3. Temperature dependence of the change  $\Delta n$  in refractive index in NiO.

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