

平成21年 5月29日現在

研究種目：科学研究費補助金（若手研究（スタートアップ））

研究期間：2007年度～2008年度

課題番号：19840026

研究課題名（和文） 新奇なマルチフェロイック酸化物の時間分解分光

研究課題名（英文） Time-resolved spectroscopic study of exotic multiferroic oxides

研究代表者

K・R MAVANI

京都大学・物質－細胞統合システム拠点・特定研究員(WPI)

研究者番号：10452260

研究成果の概要：

We synthesized different exotic oxide thin film and polycrystalline pellets. We observed electromagnons in CoCr_2O_4 thin film using terahertz time-domain spectroscopy, which is the first such observation for spinel multiferroic. Raman spectroscopy shows transit phonon mode on CoCr_2O_4 . Photoinduced effect has been observed in a manganite thin film using visible pump terahertz probe spectroscopy.

交付額

(金額単位：円)

	直接経費	間接経費	合計
平成19年度	1,260,000	0	1,260,000
平成20年度	1,310,000	393,000	1,703,000
総計	2,570,000	393,000	2,963,000

研究分野：数物系科学

科研費の分科・細目：物性I・4302

キーワード：強相関電子系、マルチフェロイック、テラヘルツ分光、ラマン分光

1. 研究開始当初の背景

Multiferroic materials are promising for future generation of devices and applications. These oxides show strong correlations between magnetic and electrical properties, where a magnetic field can influence both, the magnetic and electrical properties and similarly, the electric field can influence the same. Also strain can modify the magnetic and electrical properties. The CoCr_2O_4 is the first oxide found to show correlated and spontaneous ferroelectric and

ferromagnetic transitions due to its conical spin structure. The AB_2O_4 -type frustrated magnetic ferroelectrics can have complex spin-structures or magnons. The electromagnetic waves (THz/light) when passed through such materials, may sense the angular/spiral configuration of the spins. Thus, terahertz spectroscopy is promising tool to probe this phenomenon. Previously, terahertz techniques were employed to study high temperature superconductors and manganese oxides. Thus, terahertz spectroscopy has been proved promising for studying oxides.

2. 研究の目的

Exotic oxides like CoCr_2O_4 and LuFe_2O_4 were planned to synthesize using solid state reaction method and pulsed laser deposition method. These oxides were planned to study using Terahertz spectroscopic techniques. The aim of the study was to observe the charge dynamics and explore the possibilities of electromagnon excitations and terahertz emission from these oxides, besides studying their basic structural and magnetic properties.

3. 研究の方法

For synthesis of polycrystalline and thin film samples, we used the following methods.

- 1) Solid state reaction method using furnaces to make pellets of multiferroic materials.
- 2) Pulsed Laser Deposition method for making thin films.

The following methods were used to study the exotic materials.

- 3) Temperature dependent terahertz time-domain spectroscopy
- 4) Visible pump and terahertz probe spectroscopy for photoinduced effect in a manganite.
- 5) Temperature dependent Raman spectroscopy to study polycrystalline CoCr_2O_4 sample.
- 6) SQUID magnetometer for magnetization measurements.
- 7) AFM microscope for imaging the thin film surface.

4. 研究成果

1) Electromagnons in CoCr_2O_4 thin film:

The thin film of multiferroic CoCr_2O_4 was studied by terahertz time-domain spectroscopy in the temperature range of 10 - 160 K, where consecutive magnetic phase transitions and ferroelectric transition manifest in this Heisenberg ferrimagnet. We have reported the first observation of electromagnons in spinel structure and their systematic

temperature dependence. The frequency dependent complex dielectric constants fit to multiple Lorentz oscillators. The frequency- and temperature-dependent results suggest the interactions of electromagnons with the optical phonons. There are large dielectric anomalies corresponding to the magnetic transition temperatures, which indicate strong magnetodielectric coupling in this thin film.

The following figure shows the terahertz spectra with multiple electromagnons at three typical temperatures.

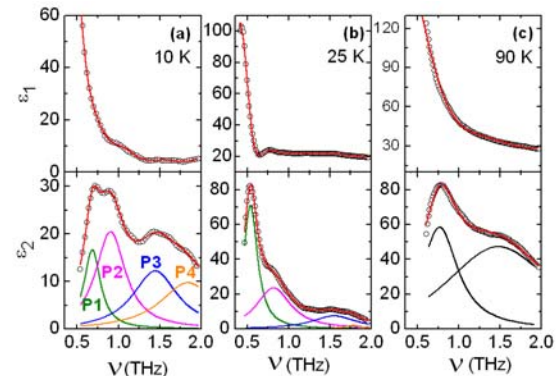


Fig. 1 (a-c): Deconvolution of the fits of ϵ_1 and ϵ_2 data to the multiple Lorentz oscillator model at three different temperatures. The four oscillators are shown by distinct colors and denotations. Above T_s , the P1-P2 and P3-P4 merge as shown by black lines for 90 K.

2) Raman spectroscopy of CoCr_2O_4 polycrystalline sample:

Temperature dependent Raman spectroscopy of polycrystalline sample of multiferroic CoCr_2O_4 shows that there are temperature dependent transit phonon modes at 500 nm which appear around magnetic transitions.

3) Ultra-fast photoinduced charge dynamics in manganite thin film observed by visible pump and terahertz probe spectroscopy:

The 800 nm laser pulse, with 150 fs pulse-width and $\sim 500 \mu\text{J}$ energy per pulse, induced picosecond charge-dynamics in $\text{Pr}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ manganite. As shown in the figure 2, $\text{Pr}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ film shows ultra-fast photoinduced dynamics. These charge dynamics are temperature dependent. Above and below ferromagnetic transition,

the charge-dynamics show slower and faster response to the laser irradiation on the thin film. The change in terahertz field shows the time-dependent changes in the charge dynamics at different temperatures. We also found that these dynamics correspond well with the temperature dependent magnetization of the thin film.

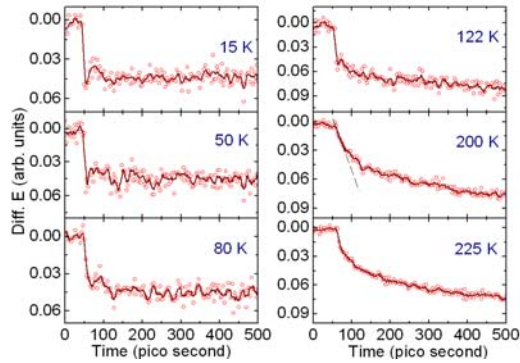
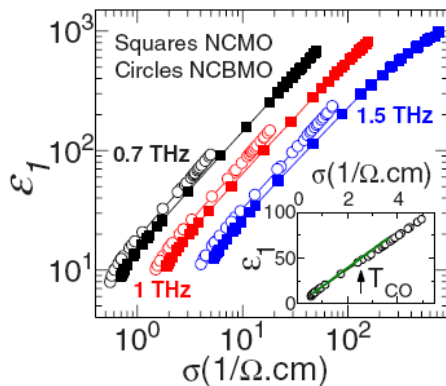


Fig. 2: Normalized photoinduced change in the main THz peak [Diff. E= (E_{pump-on} - E_{pump-off}) / E_{pump-off}] vs. time at different temperatures.

4) Cation disorder effect and scaling of optical conductivity in manganite thin films:

Fig. 3: Scaling of optical parameters at



different temperatures. The inset figure shows deviation from scaling for NCMO manganite at charge-ordering temperature. [Ref. K. R. Mavani *et al.*, Appl. Phys. Lett. 93, 231908 (2008).]

We have explored the low energy charge dynamics of charge-ordered Nd_{0.5}Ca_{0.5}MnO₃ (NCMO) and Ba-doped Nd_{0.5}Ca_{0.48}Ba_{0.02}MnO₃ (NCBMO) manganite thin films by recording the complex optical spectra using terahertz time-domain spectroscopy. The

extracted frequency- and temperature-dependent optical conductivity (σ_I) and dielectric constant (ϵ_I) show distinct features at the Néel's ordering and the charge-ordering temperatures (T_N and T_{CO}). We found that the Ba-doping induced cation disorder effects on σ_I and ϵ_I are more pronounced above T_{CO} , but these effects are subtle in a large temperature region below T_{CO} . Both, ϵ_I and σ_I , increase as a function of temperature and form a scaling relationship, $\epsilon_I \propto \sigma_I$ (Fig. 3).

5. 主な発表論文等

(研究代表者、研究分担者及び連携研究者には下線)

[雑誌論文] (計 1 件)

1. K. R. Mavani, M. Nagai, D. S. Rana, H. Yada, I. Kawayama, M. Tonouchi and K. Tanaka

Applied Physics Letters 93 (2008) 231908,

Effects of disorder and scaling of optical conductivity in Nd_{0.5}Ca_{0.5-x}Ba_xMnO₃ (x = 0 and 0.02) thin films as observed by terahertz time-domain spectroscopy”

[学会発表] (計 6 件)

1. K. R. Mavani, D. S. Rana, M. Nagai, M. Tonouchi, K. Tanaka

Japanese Physical Society Meeting at Rikkyou University, Japan, March 27-30, 2009.

“Ultra-Fast Photoinduced Effects on the Charge-Dynamics of Pr_{0.5}Sr_{0.5}MnO₃ Manganite”

2. K. R. Mavani, M. Nagai, D. S. Rana, M. Tonouchi, K. Tanaka

International Workshop on Optical Terahertz Science and Technology, Santa Barbara, United States, March 7-11, 2009.

“Large Magnetodielectric Effect Observed In Multiferroic CoCr204

- Thin Film Using Terahertz Time-Domain Spectroscopy”
3. K. R. Mavani, M. Nagai, D. S. Rana, M. Tonouchi, K. Tanaka.

Japanese Physical Society meeting at Iwate University, Japan, during September 20–23, 2008.

Poster presentation: “Influence of ferromagnetic ordering and relaxation dynamics in CoCr_2O_4 multiferroic thin film as studied by terahertz time-domain spectroscopy”
 4. K. R. Mavani, M. Nagai, D. S. Rana, M. Tonouchi, K. Tanaka.

The 8th International conference on excitonic processes in condensed matter (EXCON’ 08) at Kyoto University, Japan, during June 22–27, 2008.

Oral Presentation: “Large drop in dielectric constant at ferromagnetic transition in CoCr_2O_4 thin film as studied by terahertz time-domain spectroscopy”
 5. K. R. Mavani, M. Nagai, D. S. Rana, H. Yada, I. Kawayama, M. Tonouchi and K. Tanaka

American Physical Society meeting (APS, Spring meeting 2008) at New Orleans, USA during March 9–14, 2008.

Oral Presentation: “Scaling of optical conductivity and disorder effect in half-doped manganites $\text{Nd}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ and $\text{Nd}_{0.5}\text{Ca}_{0.48}\text{Ba}_{0.02}\text{MnO}_3$ thin films as probed by Terahertz time-domain spectroscopy.
 6. K. R. Mavani, M. Nagai, M. Shirai, K. Tanaka, D. S. Rana, I. Kawayama, M. Tonouchi

American Physical Society meeting (APS, Spring meeting 2008) at New Orleans, USA during March 9–14, 2008.

Poster presentation: “Large drop in dielectric constant at ferromagnetic transition in CoCr_2O_4 thin film as studied by terahertz time-domain spectroscopy”

6. 研究組織

(1) 研究代表者 K. R. Mavani

(2) 研究分担者 なし

(3) 連携研究者 なし