Ultrafast dynamics in multiferroics HoMnO$_3$ revealed by fs spectroscopy

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Outline

- Introduction of femtosecond (fs) laser pulses
- Ultrafast dynamics in multiferroics HoMnO$_3$
- Summary I
- High-$T_c$ superconductor YBa$_2$Cu$_3$O$_7$ nanodots
- Summary II
What is the ultrashort pulse?

- $\sim 10^{-6}$ s
- $\sim 10^{-9}$ s
- $\sim 10^{-12}$ s
- $\sim 10^{-15}$ s
Introduction of fs laser pulses

Timescales

- 10 fs light pulse
- Computer clock cycle
- Camera flash
- 1 minute
- One month
- Age of pyramids
- Human existence
- Age of universe

1 femtosecond
1 picosecond

Time (seconds)

10^{-15} 10^{-12} 10^{-9} 10^{-6} 10^{-3} 10^0 10^3 10^6 10^9 10^{12} 10^{15} 10^{18}

a pulse : 1 minute ~ 1 minute : age of universe
Introduction of fs laser pulses

Which one is true?

1 / 1 min

1 / 0.5 min

1 / 1 sec

Idea from 石訓全
Introduction of fs laser pulses

Ultrafast camera!!
Introduction of fs laser pulses

The possibility for nuclear fusion!

- Short pulse = intense peak power
  \[100 \text{ mJ}, 100 \text{ fs} = 1 \text{ TW}\]
  \[10^{18} \text{ W/cm}^2 @ \phi = 10 \mu \text{m} (10^{10} \text{ V/cm})\]
Introduction of fs laser pulses

The evolution of pulse width
The shorter pulse duration, the more papers!

- Intra-cavity pulse compression
- XUV excitation pulse
- Colliding pulse mode-locking
- Passive mode-locking
- Active mode-locking
- First laser (Ruby)

Year

Pulse duration (sec.)

No. of Publications

Prof. Ahmed Zewail
The 1999 Nobel Prize
in Chemistry

Prof. Theodor W. Hänsch
The 2005 Nobel Prize
in Physics
Multiferroic

- Ferromagnets (ferroelectrics) form a subset of magnetically (electrically) polarizable materials such as paramagnets and antiferromagnets (paraelectrics and antiferroelectrics)

Ultrafast dynamics in HoMnO$_3$

Multiferroic ReMnO$_3$

Hexagonal structure v.s. Orthorhombic structure


Hexagonal HoMnO$_3$

MnO$_5$ bipyramids form a layered structure on $a$-$b$ plane.

$T_C = 875$ K  $P_z = 5.6$ $\mu$ C cm$^{-2}$

$T_N = 76$ K  $T_{SR} = 33$ K  $T_{Ho} = 5$ K

Coexistence between FE and AFM

Ultrafast dynamics in HoMnO$_3$

Magnetoelectric coupling effect on hexagonal HoMnO$_3$

Dielectric constant  
Heat capacity  
Lattice constant

Ultrafast dynamics in HoMnO$_3$

Optical properties of hexagonal HoMnO$_3$

Transmittance and reflectance measurements were performed using a Fourier transform spectrometer in a frequency range from 10 to 45000 cm$^{-1}$ (1.2 meV to 5 eV). A 1.7 eV absorption peak comes from $d \rightarrow d$ transitions. ~0.15 eV blueshift as decreasing temperature associates with the magnetic phase transition.

Ultrafast dynamics in HoMnO₃

Optical properties of hexagonal HoMnO₃

Rare-earth: Gd, Tb, Dy, Ho

Ultrafast dynamics in HoMnO$_3$

Crystal structure and magnetic property

Out of plane : $c$-axis
In plane : $ab$-axis

$T_N = 76$ K  $T_{SR} = 33$ K  $T_{Ho} = 5$ K

HMO(002)  HMO(004)  HMO(006)
Ultrafast dynamics in HoMnO$_3$

Pump-probe and optical spectroscopy

Tunable photon energy from 1.52 to 1.69 eV
Ultrafast dynamics in HoMnO$_3$

Temperature-dependent transient reflectivity change ($\Delta R/R$)

Wavelength: 800 nm

Wavelength: 770 nm

Wavelength: 740 nm
Ultrafast dynamics in HoMnO$_3$

Oscillation component

Strain Pulse Model

\[ \tau_{osc} \simeq \left( \frac{\lambda_{probe}}{2v_{sound}} \sqrt{n^2 - \sin^2 \theta} \right) \]

\[ \Delta R/R \text{ (arb. units)} \]

\[ \text{Delay time (ps)} \]

T=290K

800nm 740nm

LuMnO$_3$

Ultrafast dynamics in HoMnO$_3$

Charge transfer from $e_{2g}$ to $a_{1g}$ by pump pulses

\[
\begin{align*}
E_{\text{Pump energy}} & : 1.52 \text{ eV} \\
T = 290 \text{ K} & \quad \text{Room temperature} \\
T = 140 \text{ K} & \quad \text{Low temperature}
\end{align*}
\]

\[
\begin{align*}
\text{Mn}^{3+} \quad 3d \quad \text{levels} \\
\text{Observed the blueshift of energy gap!}
\end{align*}
\]

Ultrafast dynamics in HoMnO$_3$

Charge transfer from e$_{2g}$ to a$_{1g}$ by pump pulses

- Observed the blueshift of energy gap!

Graphical representation:
- Normalized amplitude of $\Delta R/R$ versus temperature (K)
- 815nm and 800nm wavelengths
- Mn$^{3+}$ 3$d$ levels
- Pump energy: 1.55 eV
- Room temperature vs. Low temperature
- $d_{3z^2-r^2}$ vs. $d_{(x^2-y^2),(xy)}$
Ultrafast dynamics in HoMnO$_3$

Charge transfer from $e_{2g}$ to $a_{1g}$ by pump pulses

![Graph showing normalized amplitude of $\Delta R/R$ vs. temperature (K) for 815nm and 800nm wavelengths, with $T_0=117$ K, pump energy 1.55 eV, and observed blueshift of energy gap.]
Ultrafast dynamics in HoMnO$_3$

Charge transfer from $e_{2g}$ to $a_{1g}$ by pump pulses

Mn$^{3+}$ 3$d$ levels

Pump energy : 1.68 eV

Room temperature Low temperature

Temperature (K)

Energy gap $E_{dd}$ (eV)

Temperature (K)

Slope

Temperature (K)

Curie-Weiss Law

ZFC 100 Oe

H//c-axis

AFM

$1/\chi$ (Oe/emu)

$\chi$ (emu/Oe)

$T_{SR}$

$T_{Ho}$
Ultrafast dynamics in HoMnO$_3$

Charge transfer from $e_{2g}$ to $a_{1g}$ by pump pulses

Extra-blueshift comes from long-range AFM ordering!!
Ultrafast dynamics in HoMnO$_3$

Demagnetization dynamics

T=290K

T=180K

T=75K

Delay time (ps)

Temperature (K)

$\tau_m$

$\tau_c$

$T_e$

$T_l$

$T_s$
Summary

- The oscillation due to the strain pulse was clearly observed in $\Delta R/R$ by fs spectroscopy.

- A distinct blueshift of the Mn$^{3+}$ d-d optical transition comes from the appearance of AFM long-range ordering.

- The demagnetization time ($\tau_m$) in a few ps scale and its recovering time ($\tau_c$) in a few 100 ps scale were shown in the $\Delta R/R$. 
YBCO nanodots

Sample preparation:

(001) YBa$_2$Cu$_3$O$_7$ (YBCO) / (100) LaAlO$_3$

XRD

$T_c = 90.1$ K

SEM

Resistance ($\Omega$)

Temperature (K)
YBCO nanodots

Experimental setup: (spot size~110 μm)
**YBCO nanodots**

**Results** – surface morphology

YBCO nanodots

Results – structure

XRD signals of YBCO thin films at various laser fluences.
YBCO nanodots

Results – superconductivity

Fluence = 0 J/cm²

Fluence = 0.21 J/cm²

Fluence = 0.26 J/cm²

Fluence = 0.32 J/cm²

Fluence = 0.53 J/cm²

Fluence = 0.54 J/cm²

Fluence = 0.55 J/cm²

Fluence = 0.56 J/cm²
YBCO nanodots

Results – composition

EDS spectra show the composition of area 1 and area 2.

\[ \Delta T = \frac{W}{CV} \]

- \( W \approx 0.1 \text{mJ} \)
- \( C = 2.86 \times 10^6 \text{J.m}^{-3}\text{K}^{-1} \)
- \( V = 1.14 \times 10^{-14} \text{m}^3 \)

3000 K > 1897 K (Ba)

3700 K > 3345 K (Y)
The surface microstructure of YBCO thin films can be manipulated by properly controlling the fluence of the irradiating femtosecond laser. A ripple pattern was clearly observed on the surface of one YBCO thin film. The (001)-YBCO film turns into nanodot array with the superconductivity remains almost intact. Serve as a new way of engineering the material surfaces into nanometer scale structures.

Formation of nano-textured conical microstructures in titanium metal surface
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