Ultrafast dynamics in multiferroics $HoMnO_3$ revealed by fs spectroscopy

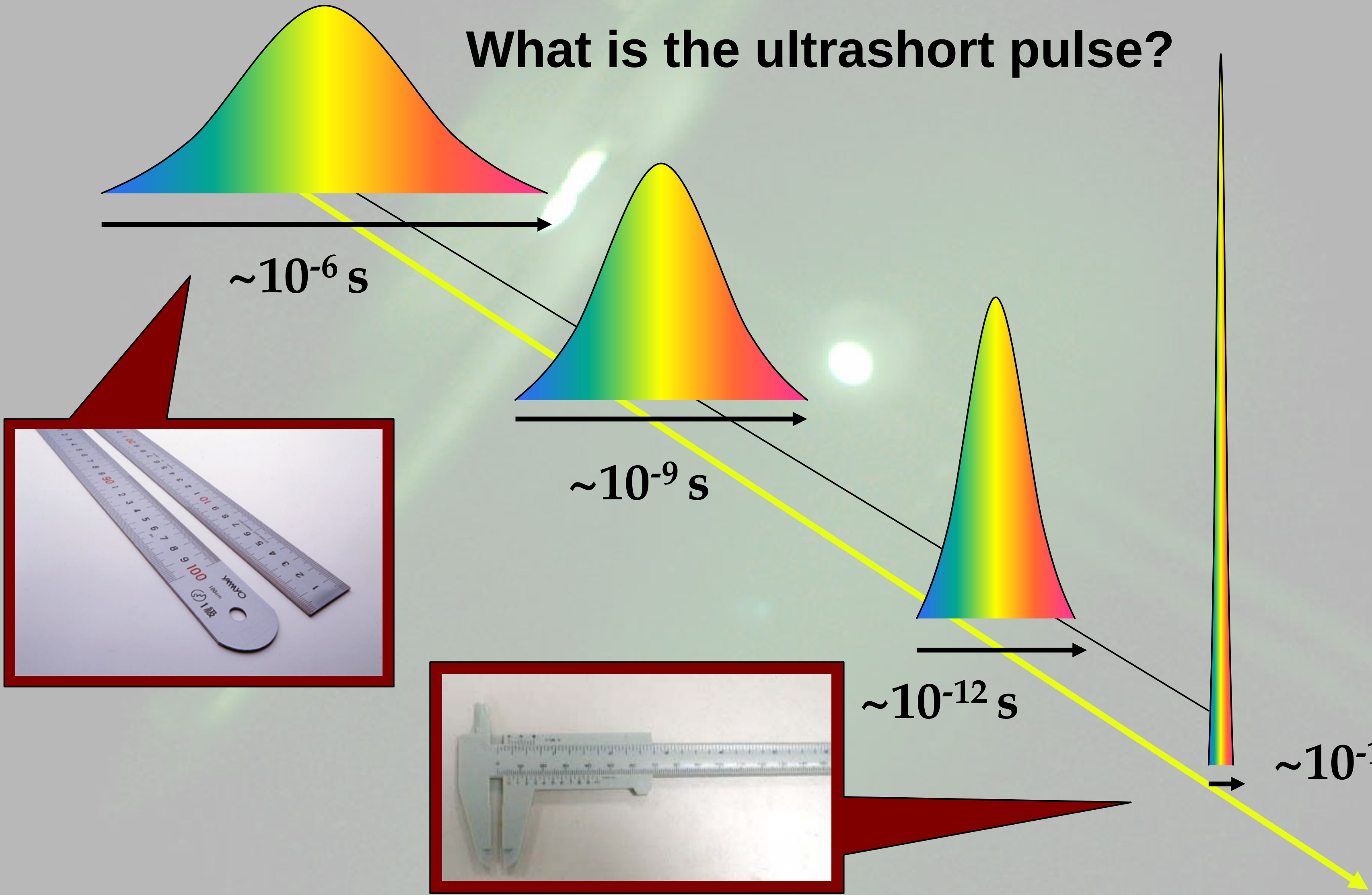
Chih Wei Luo（羅志偉）

Outline

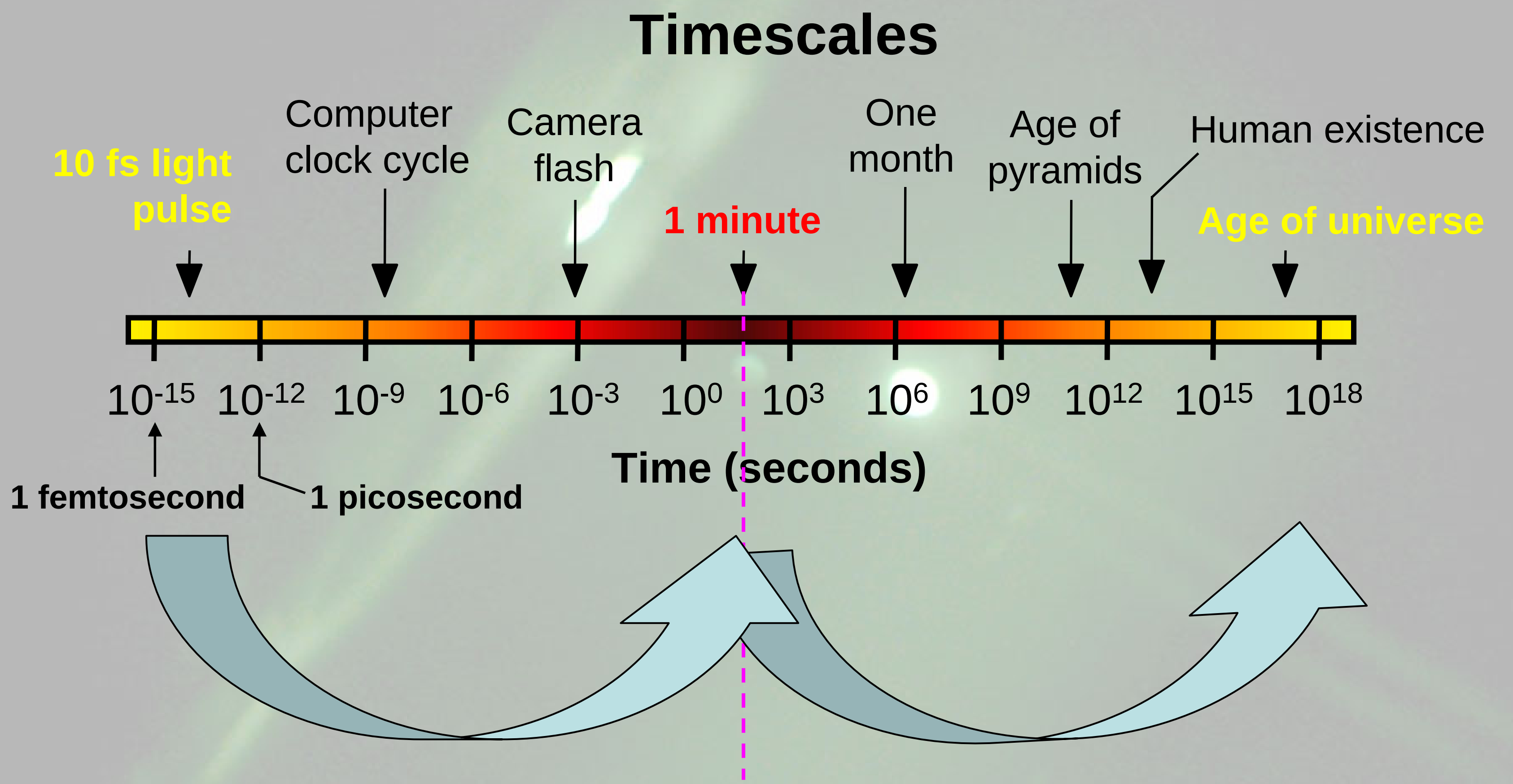
- Introduction of femtosecond (fs) laser pulses
- Ultrafast dynamics in multiferroics $HoMnO_3$
- Summary I
- High-T_c superconductor $YBa_2Cu_3O_7$ nanodots
- Summary II

Introduction of fs laser pulses

What is the ultrashort pulse?



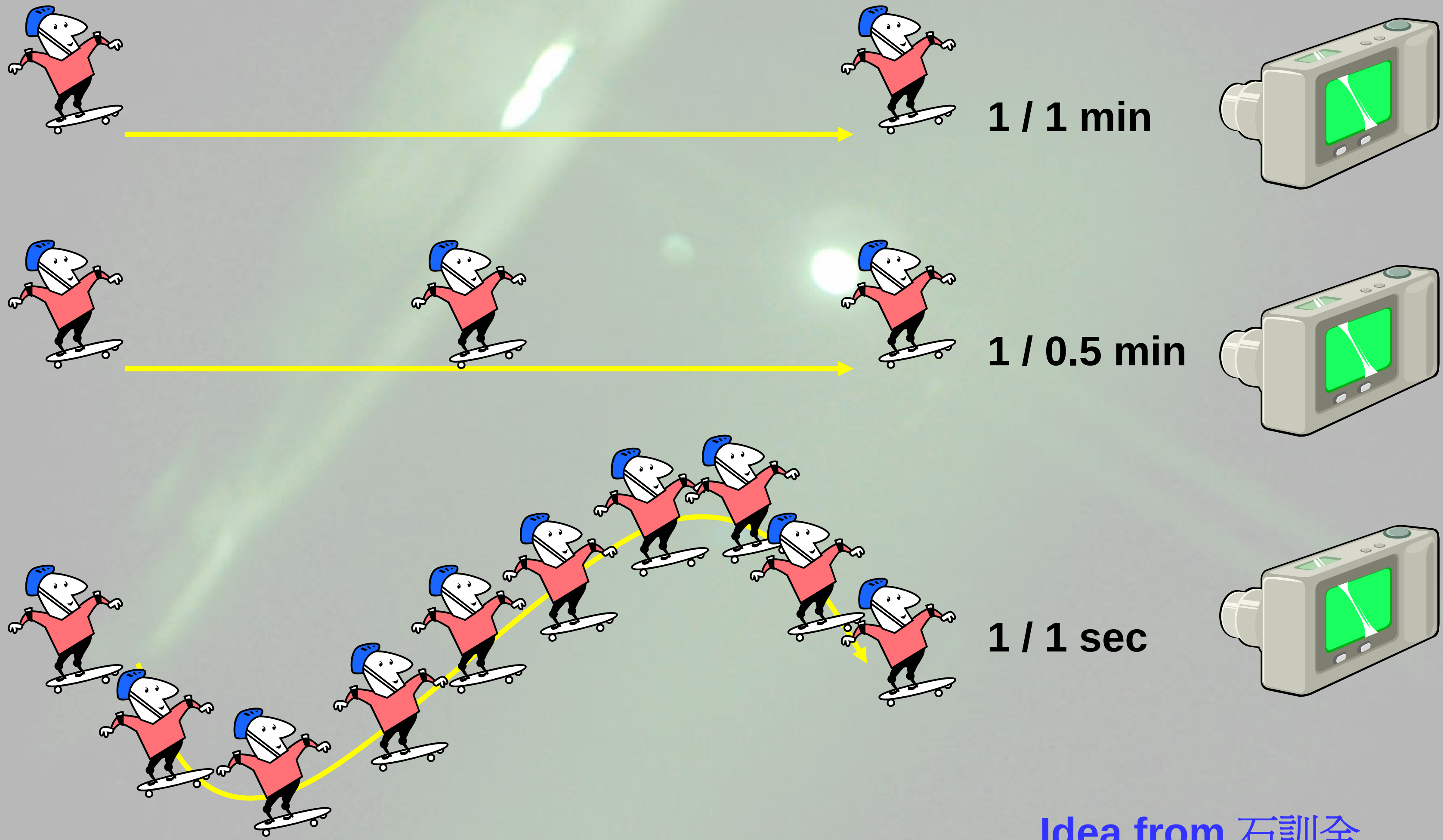
Introduction of fs laser pulses



a pulse : 1 minute ~ 1 minute : age of universe

Introduction of fs laser pulses

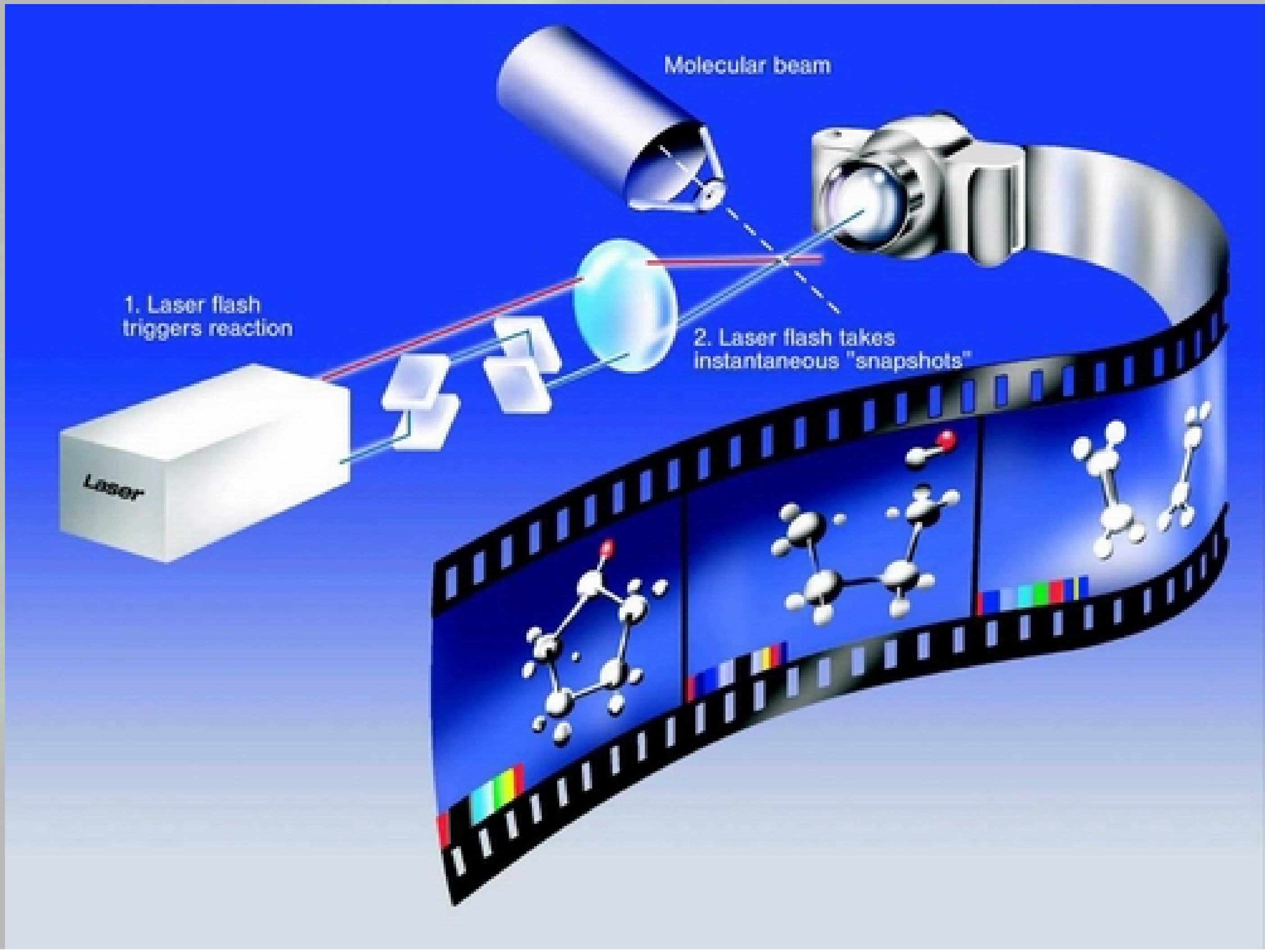
Which one is true?



Idea from 石訓全

Introduction of fs laser pulses

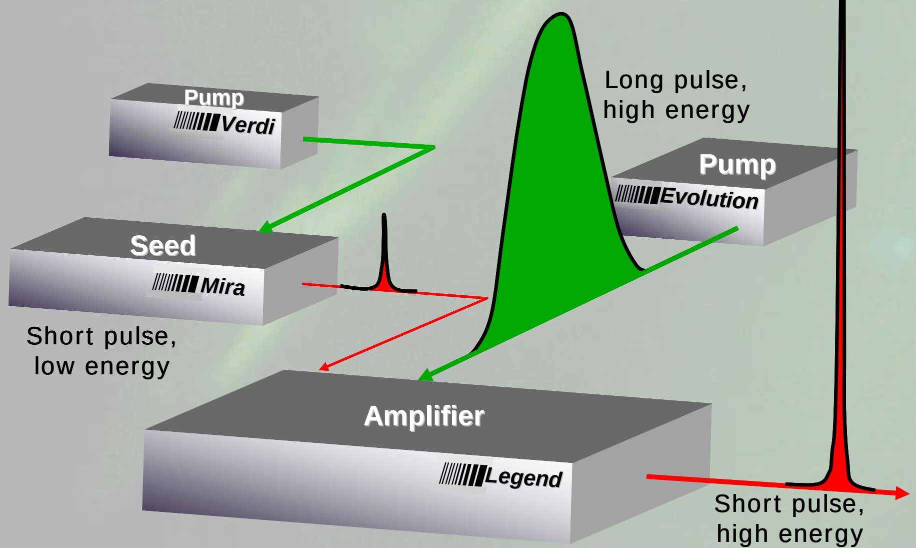
Ultrafast camera!!



Introduction of fs laser pulses

The possibility for nuclear fusion!

- Short pulse = intense peak power
  - → 100 mJ, 100 fs = 1 TW
  - → 10^{18} W/cm^2 @ $\phi = 10\ \mu$m (10^{10} V/cm)

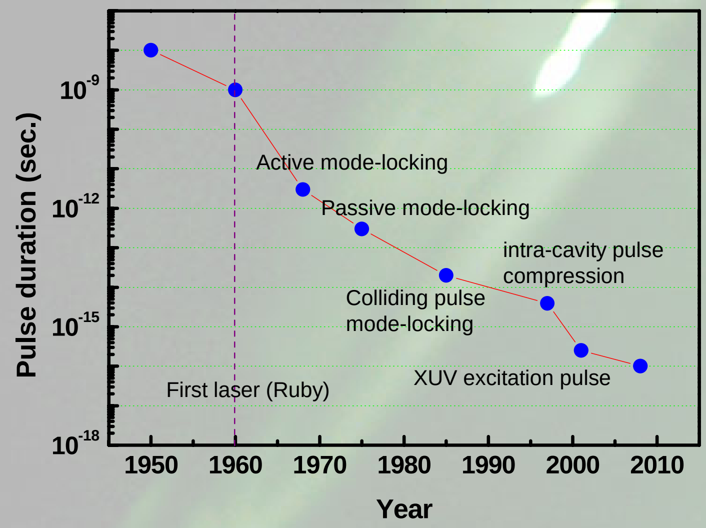


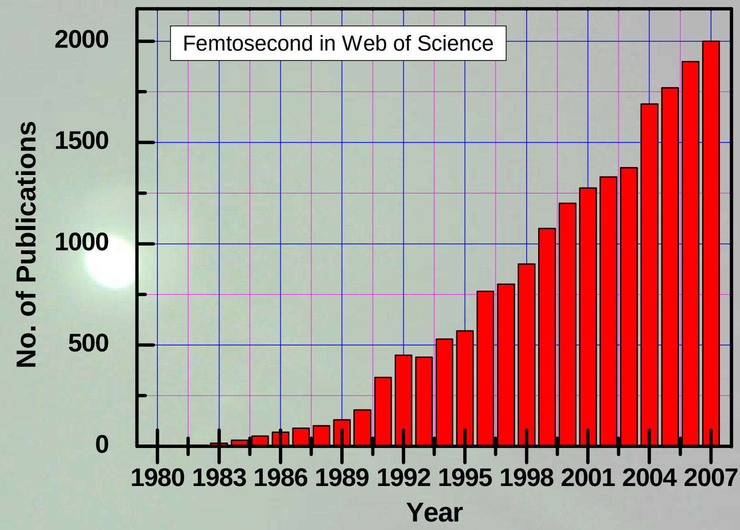
Institute of Laser Engineering
Osaka University

Introduction of fs laser pulses

The evolution of pulse width

The shorter pulse duration, the more papers!





Prof. Ahmed Zewail

The 1999 Nobel Prize in Chemistry

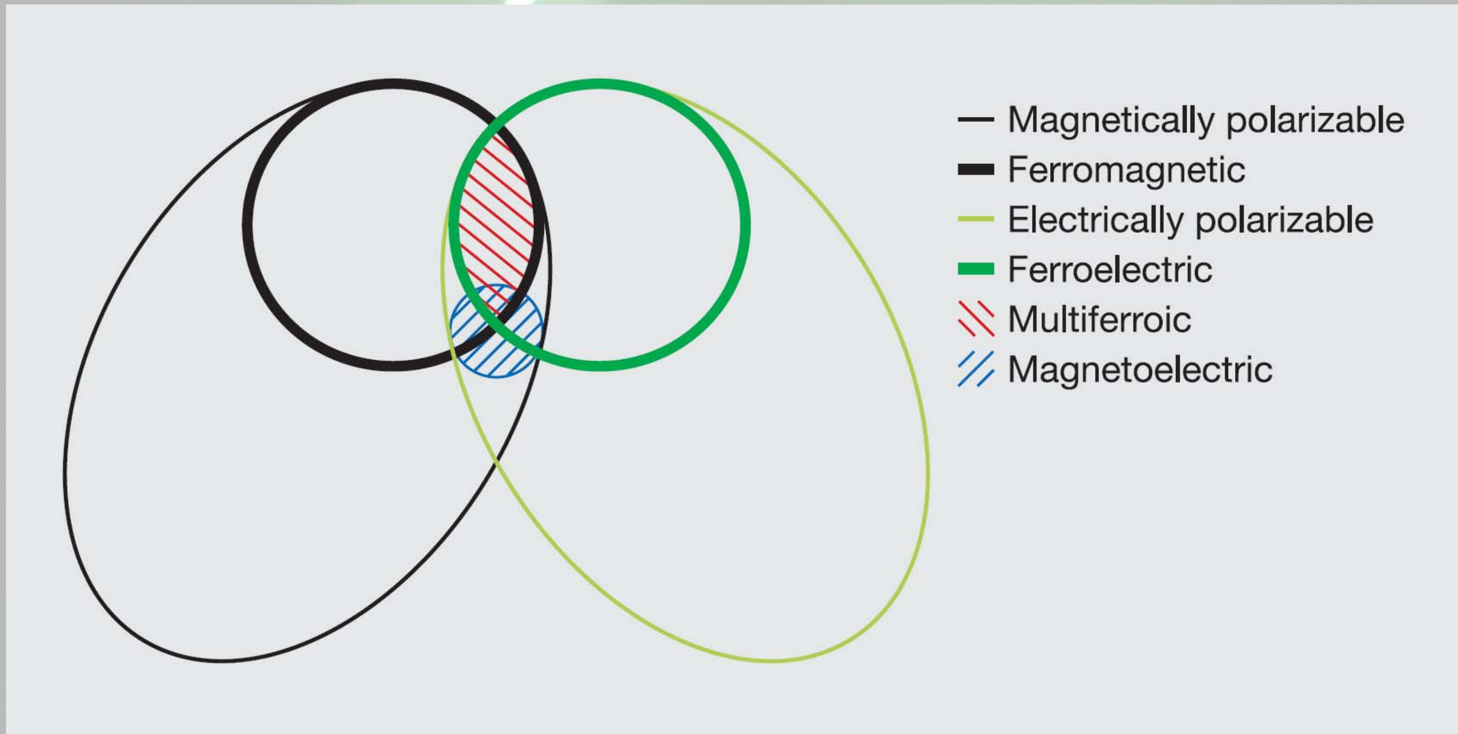
Prof. Theodor W. Hänsch

The 2005 Nobel Prize in Physics

Ultrafast dynamics in $HoMnO_3$

Multiferroic

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

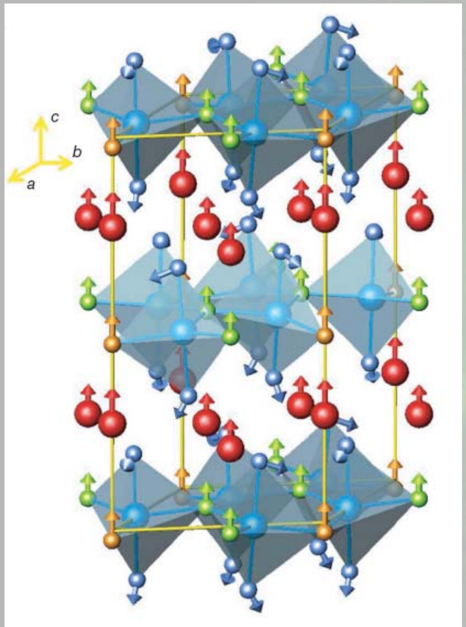


W. Eerenstein, N.D. Mathur, J.F. Scott, Nature **442**, 759 (2006).

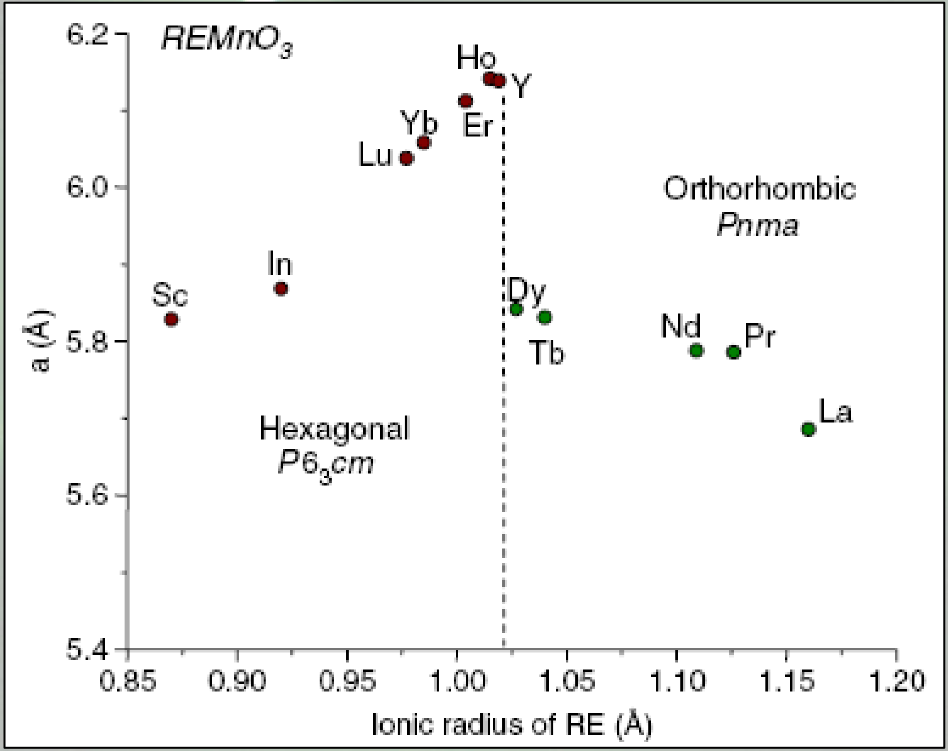
Ultrafast dynamics in $HoMnO_3$

Multiferroic *Re*MnO_3

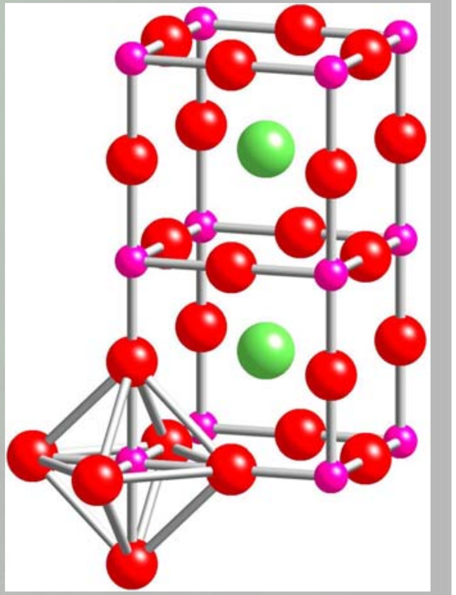
Hexagonal structure v.s. Orthorhombic structure



Seongsu Lee, et al Nature **451**,805 (2008)



W. Prellier, et al, JPCM **17**, 803 (2005)

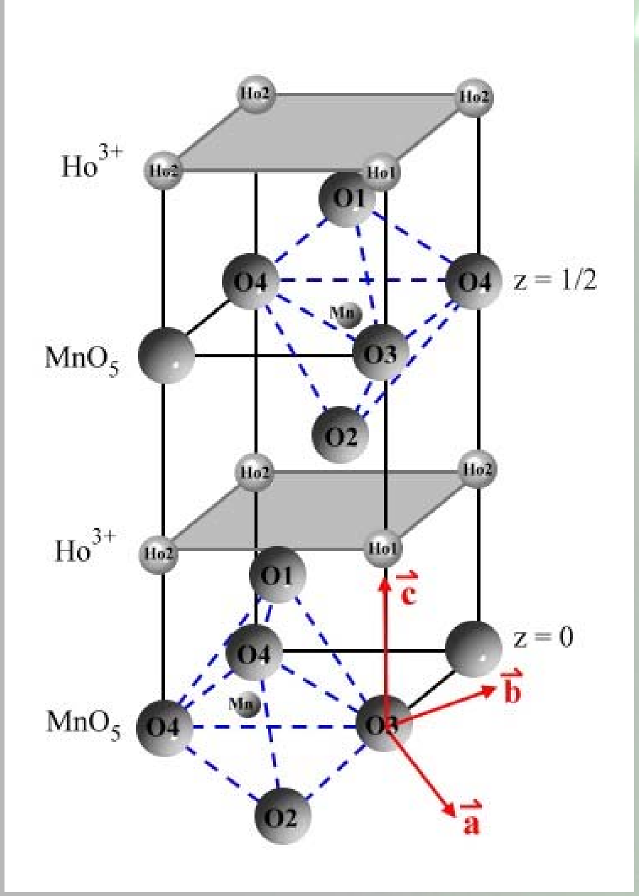


S. Satpathy, et al PRL **76** ,960 (1996)

Ultrafast dynamics in $HoMnO_3$

Hexagonal $HoMnO_3$

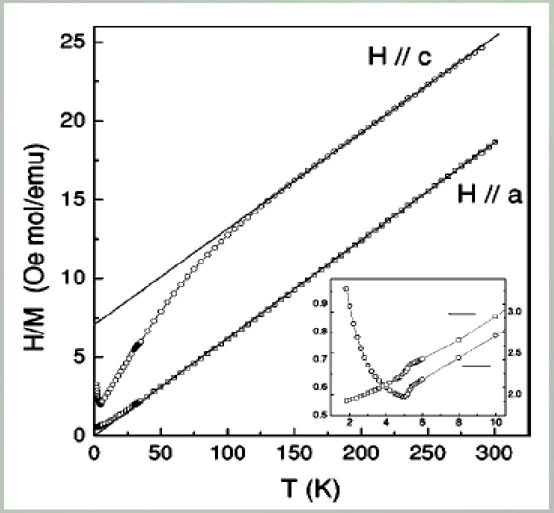
MnO_5 bipyramids form a layered structure on *a-b* plane.

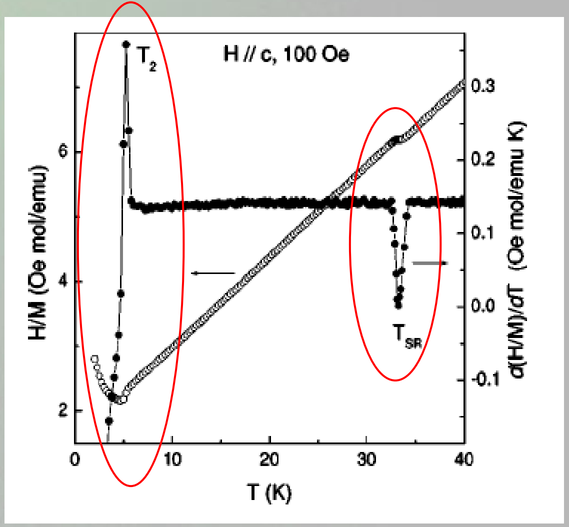


T_C= 875 K P_z= 5.6 μC cm^{-2}

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

Coexistence between FE and AFM

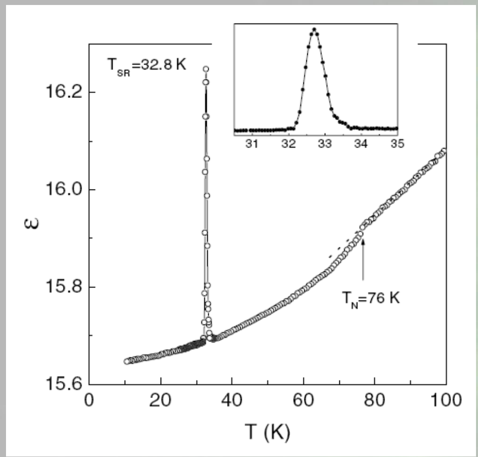




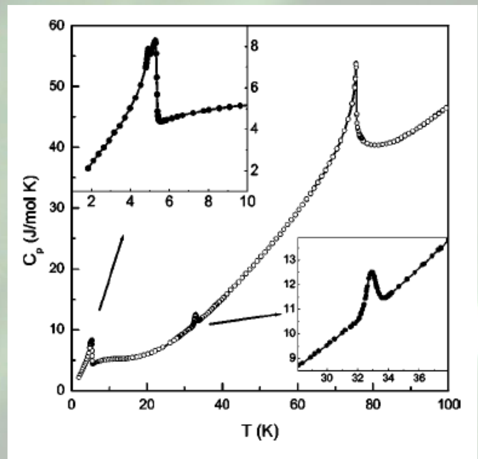
B. Lorenz, et al PRB **71** ,014438 (2005)

Ultrafast dynamics in $HoMnO_3$

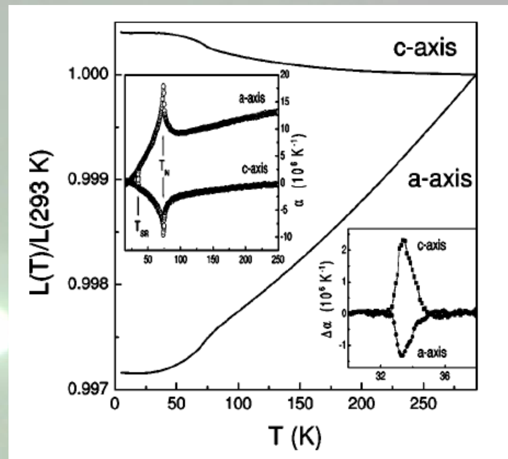
Magnetoelectric coupling effect on hexagonal $HoMnO_3$



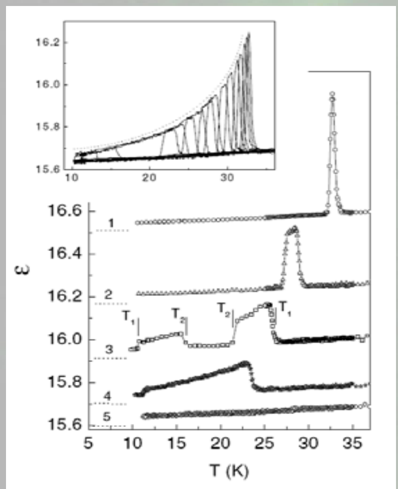
Dielectric constant

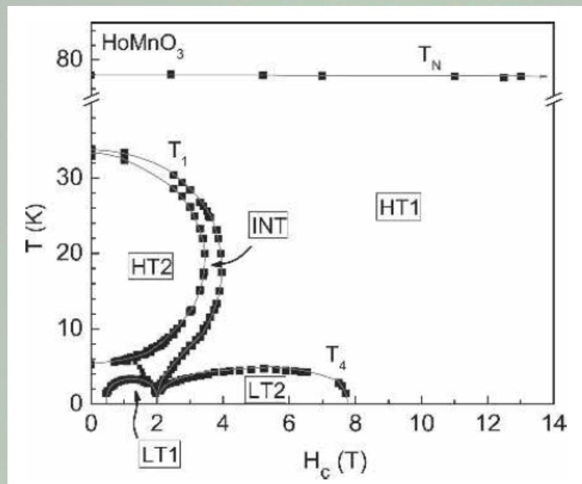


Heat capacity



Lattice constant





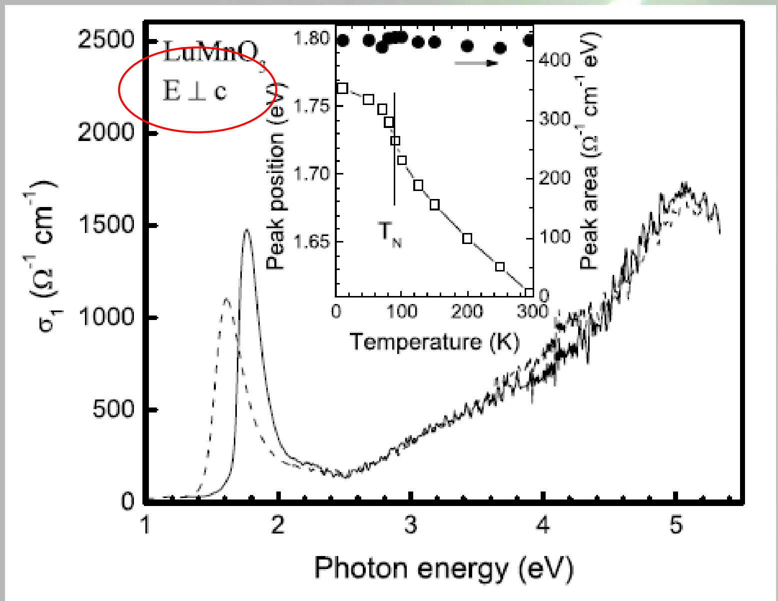
B. Lorenz, et al PRL **92** ,087204 (2004)

B. Lorenz, et al PRB **71** ,014438 (2005)

C. Dela Cruz, et al PRB **71** ,060407**R** (2005)

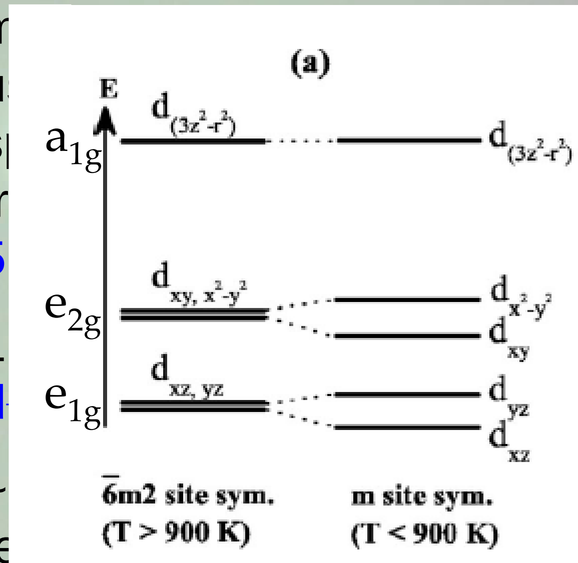
Ultrafast dynamics in $HoMnO_3$

Optical properties of hexagonal $HoMnO_3$



A. B. Souchkov, et al PRL **91** ,027203 (2003)

- Transmittance and reflectance m… …erformed u… …ransform s… …cy range fr… meV to 5…
- 1… …nes from d…
- ~… …creasing te…

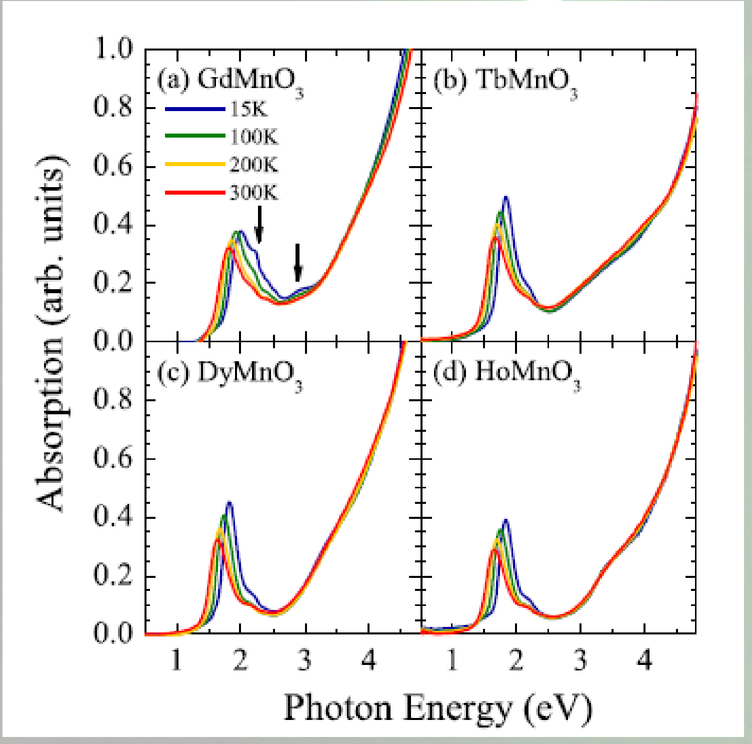


Associate with the magnetic phase transition.

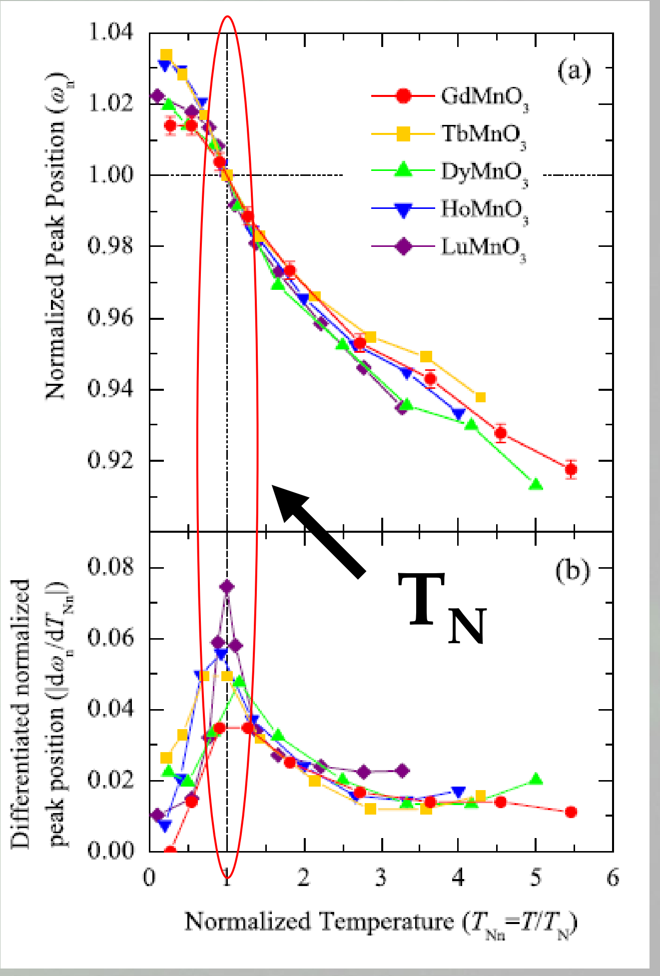
Ultrafast dynamics in $HoMnO_3$

Optical properties of hexagonal $HoMnO_3$

Rare-earth : Gd 、Tb、Dy 、Ho



Woo Seok Choi, et al PRB **78** ,054440 (2008)



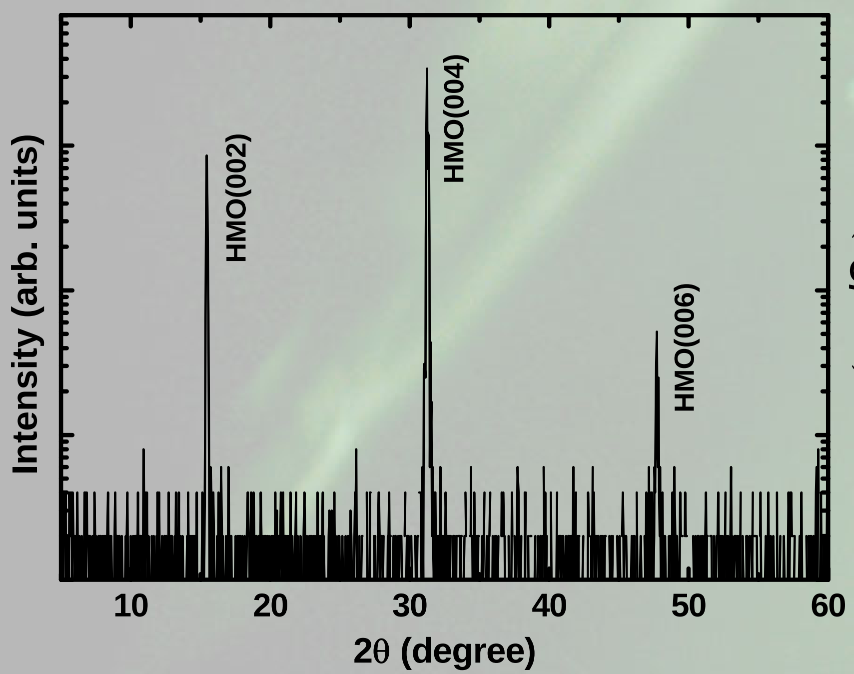
Ultrafast dynamics in $HoMnO_3$

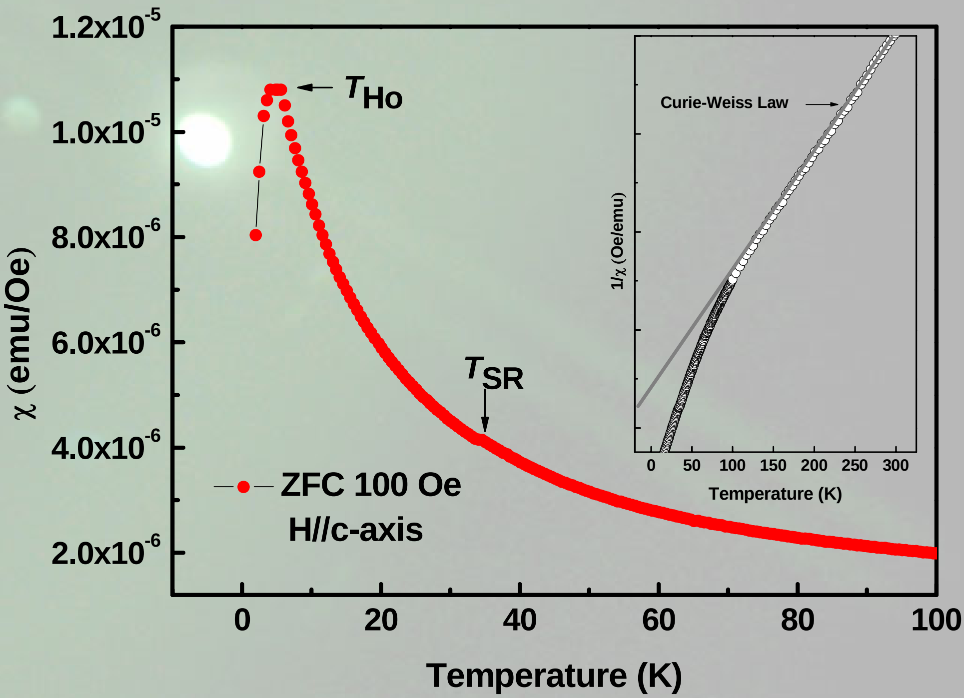
Crystal structure and magnetic property

Out of plane : *c*-axis

In plane : *ab*-axis

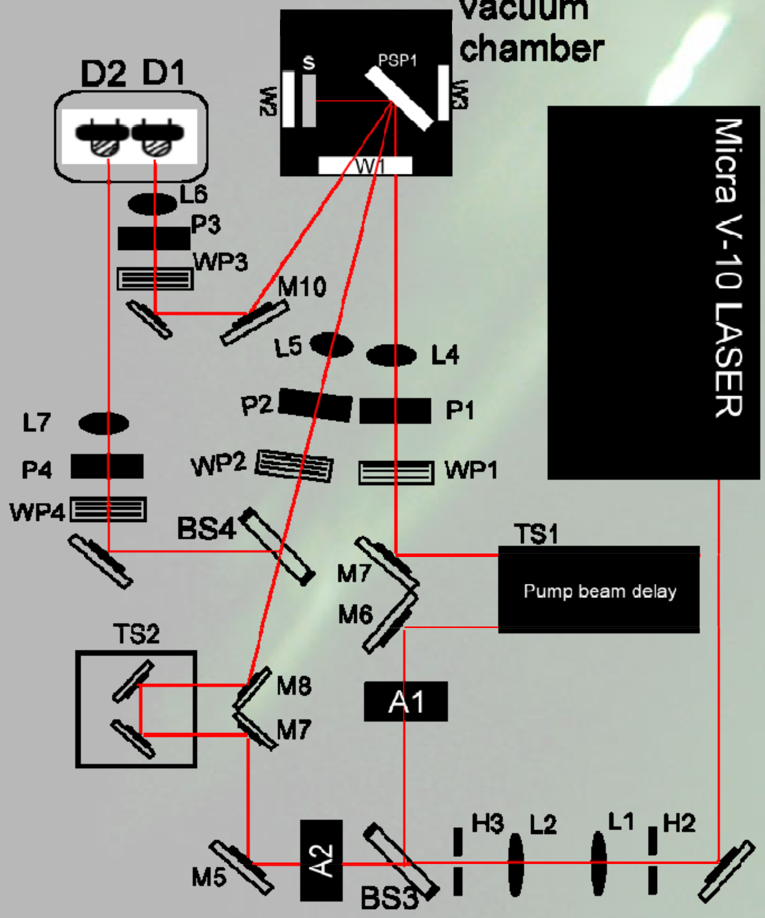
T_N= 76 K T_{SR}= 33 K T_{Ho}= 5 K



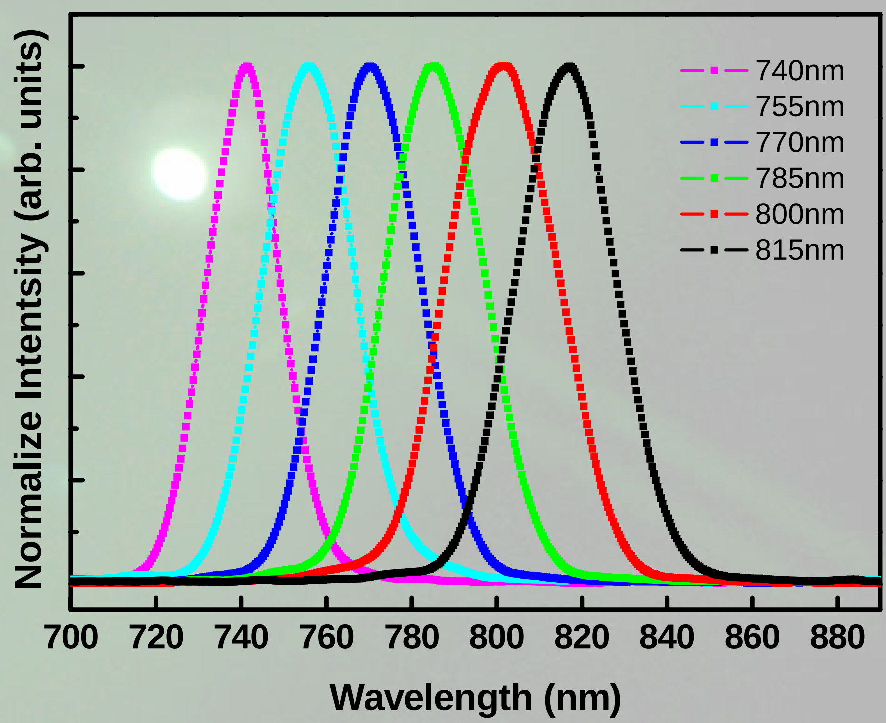


Ultrafast dynamics in $HoMnO_3$

Pump-probe and optical spectroscopy

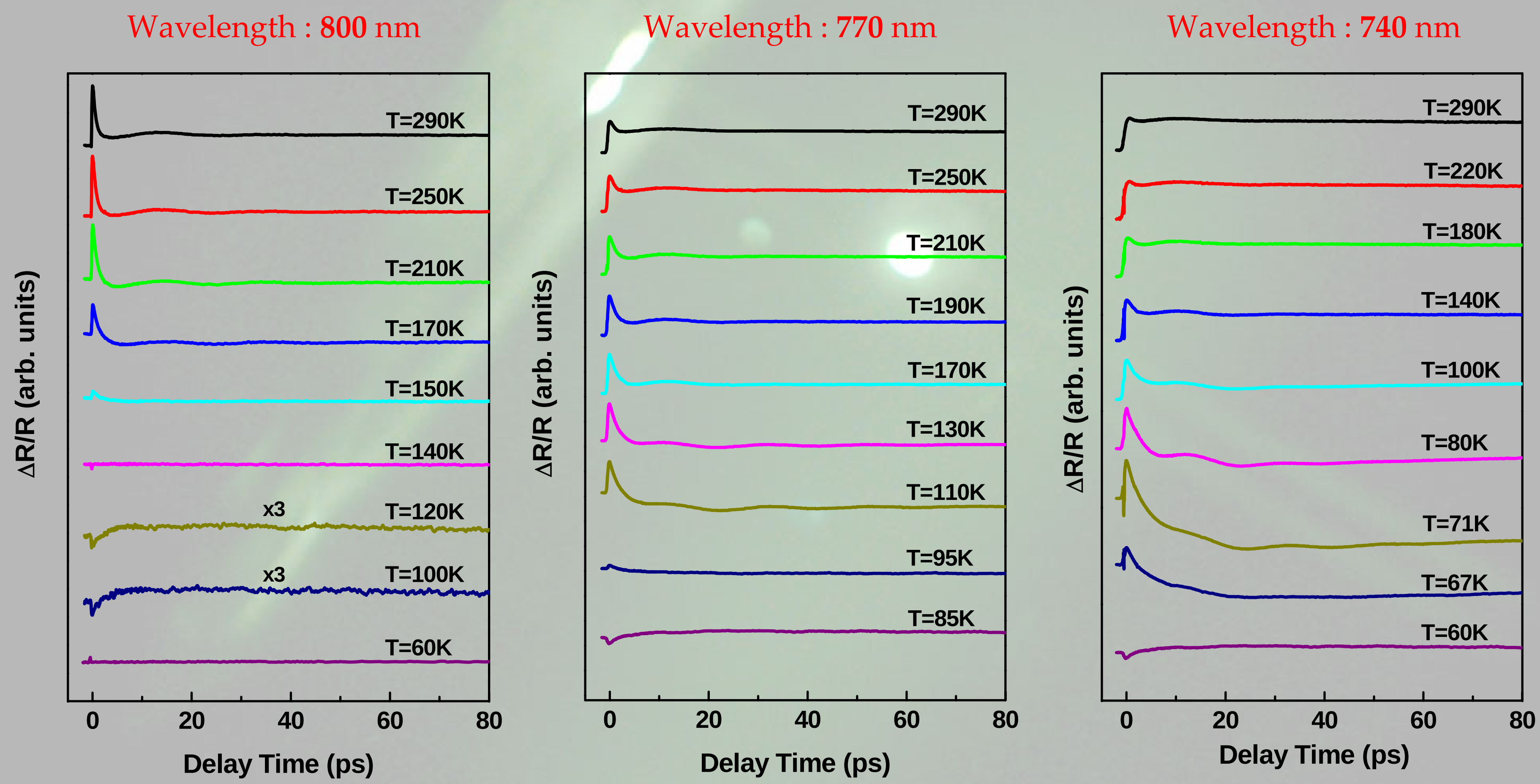


Tunable photon energy from 1.52 to 1.69 eV



Ultrafast dynamics in $HoMnO_3$

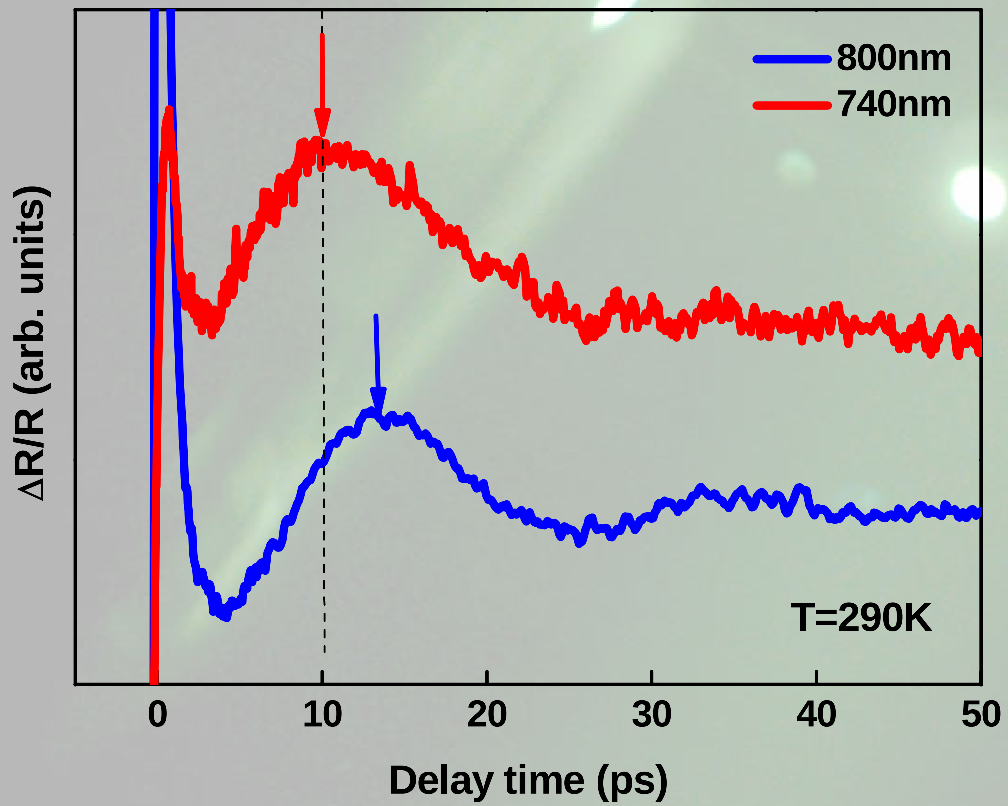
Temperature-dependent transient reflectivity change ($\Delta R/R$)

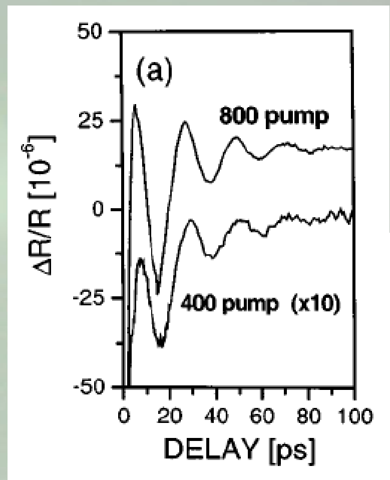


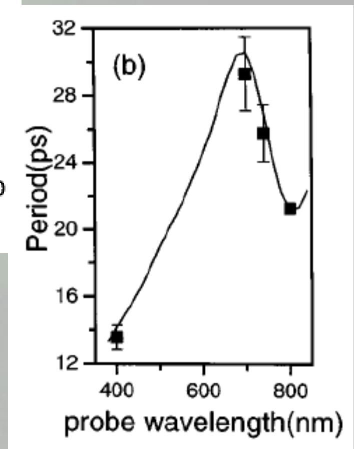
Ultrafast dynamics in $HoMnO_3$

Oscillation component

⇨ Strain Pulse Model ⇨ $\tau_{osc} \cong (\lambda_{probe} / 2\upsilon_{sound} \sqrt{n^2 - \sin^2\theta})$





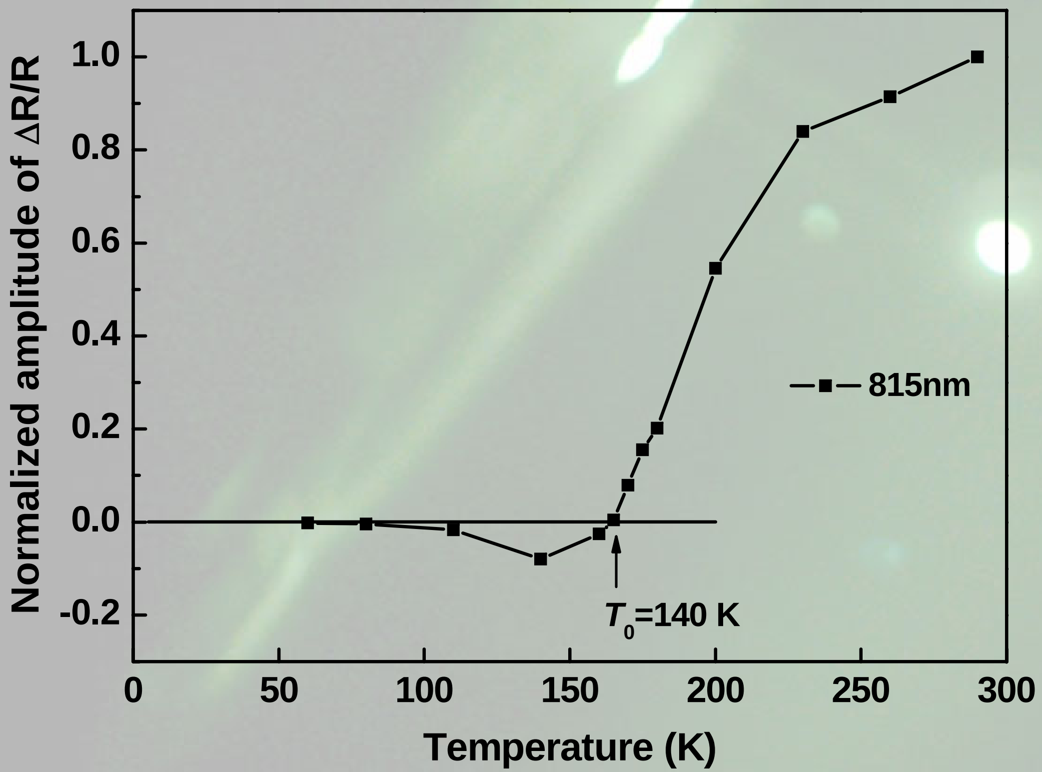


$LuMnO_3$

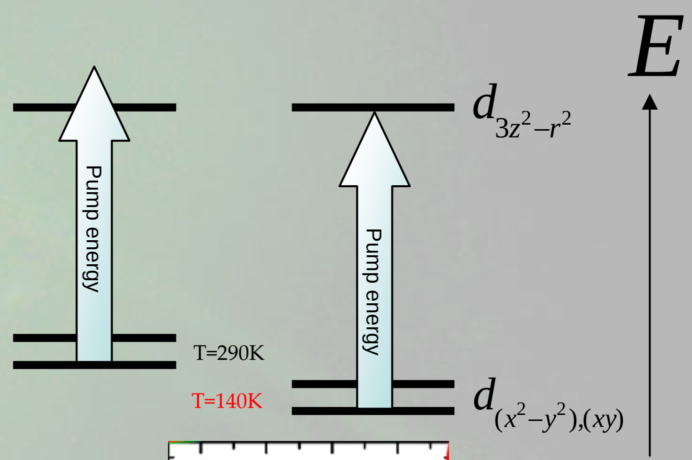
D. Lim, et al APL **83** ,4800 (2003)

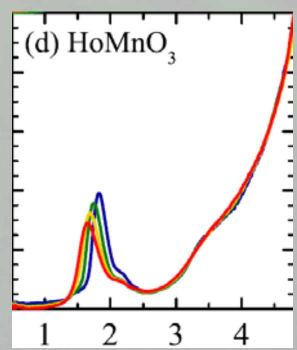
Ultrafast dynamics in $HoMnO_3$

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



Mn^{3+} 3*d* levels Pump energy :**1.52** eV



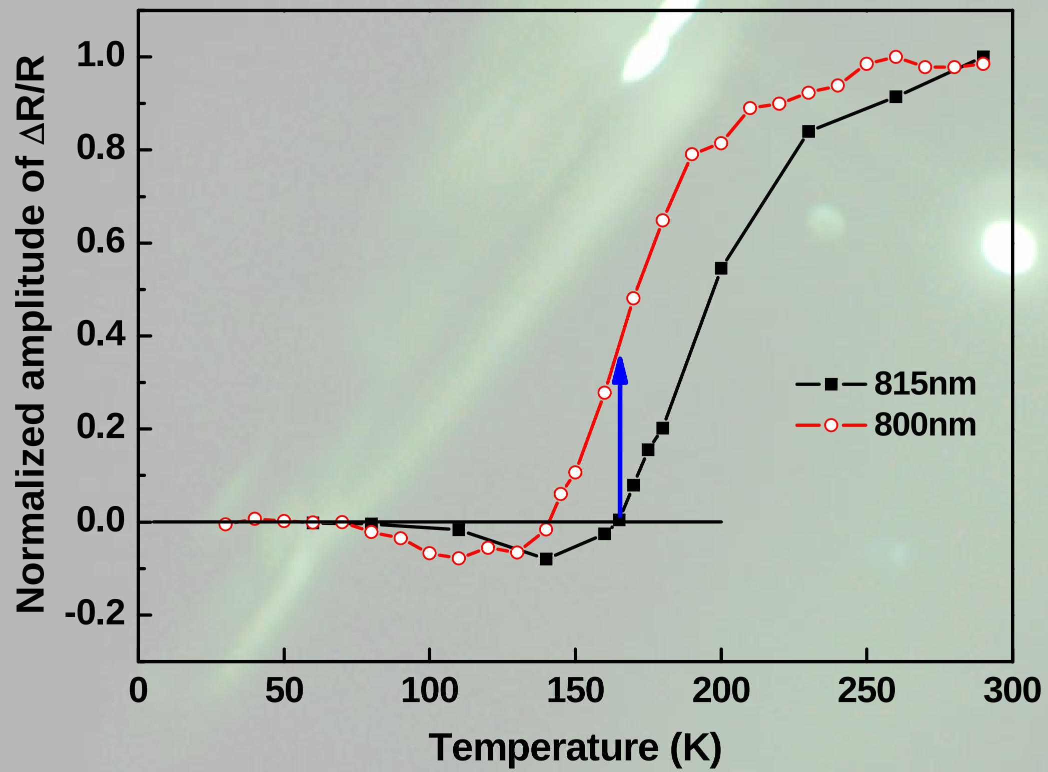


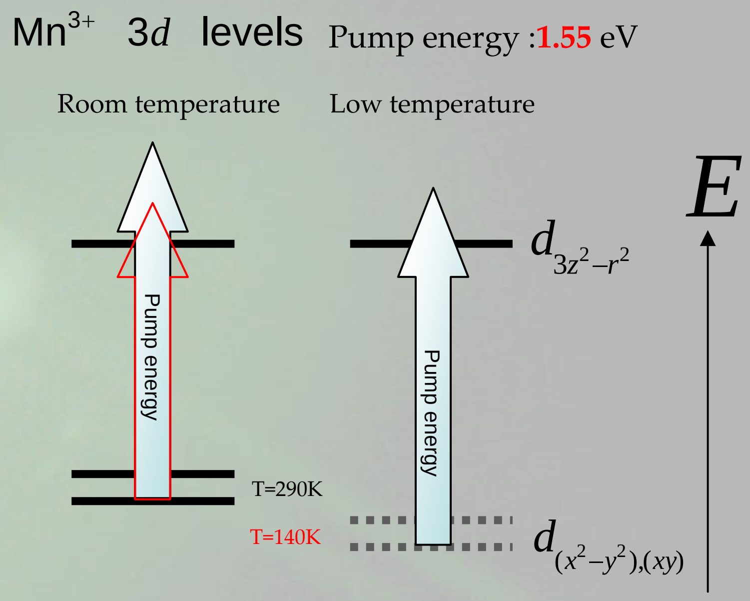
Woo Seok Choi, et al PRB **78** ,054440 (2008)

- Observed the blueshift of energy gap !

Ultrafast dynamics in $HoMnO_3$

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

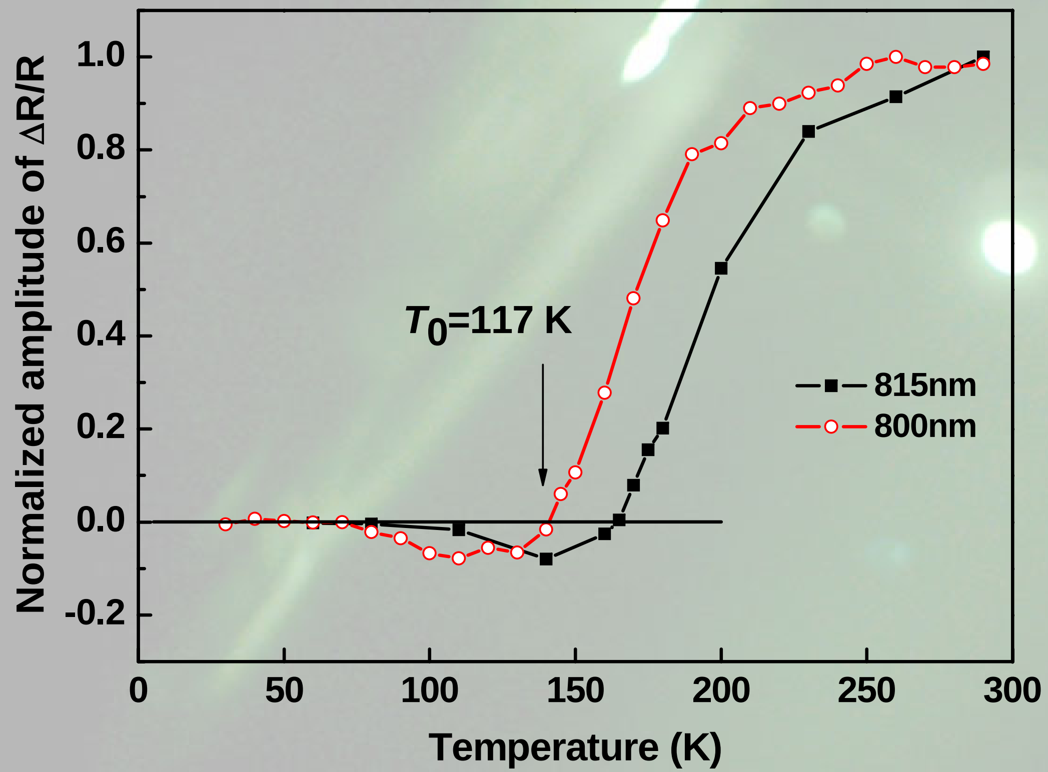


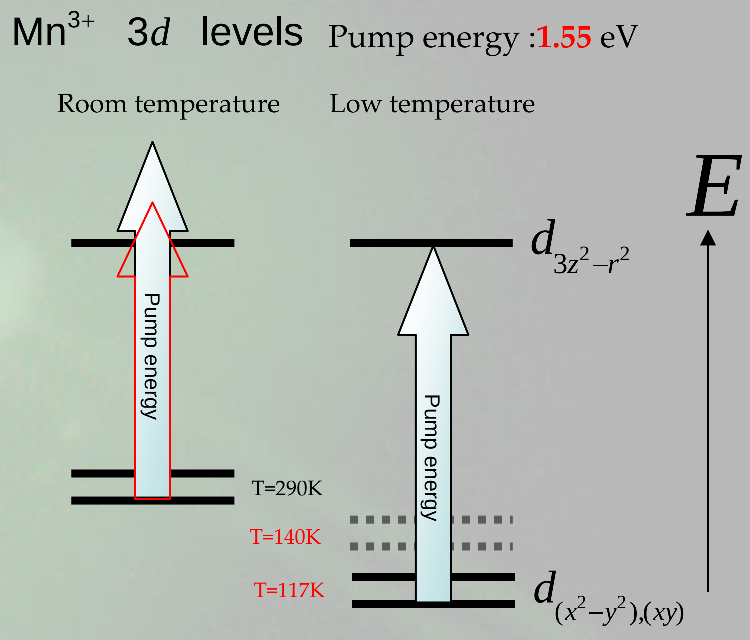


□ Observed the blueshift of energy gap !

Ultrafast dynamics in $HoMnO_3$

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

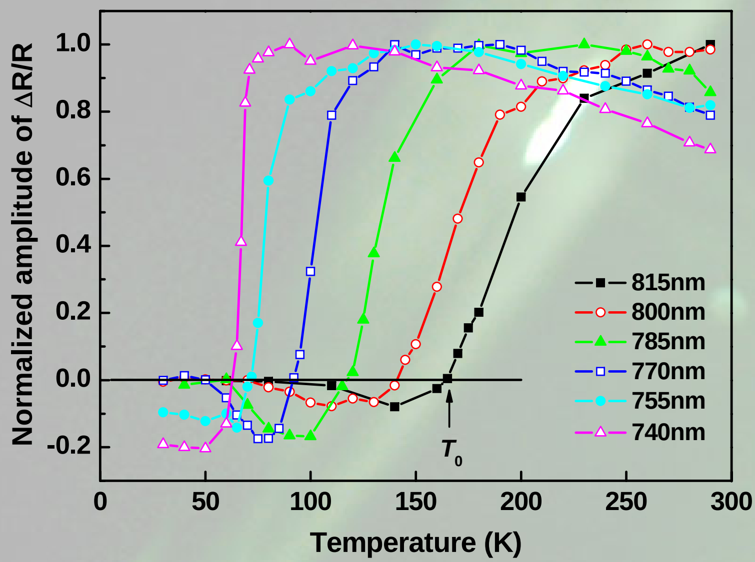


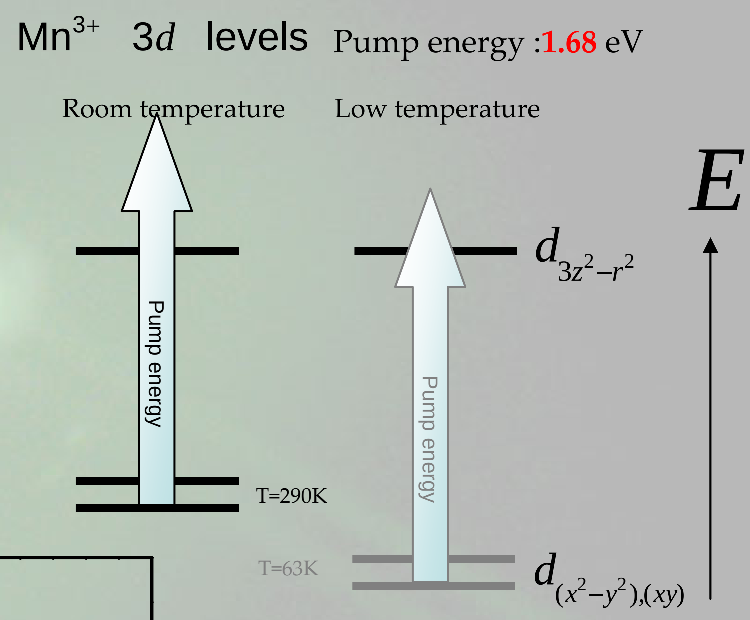


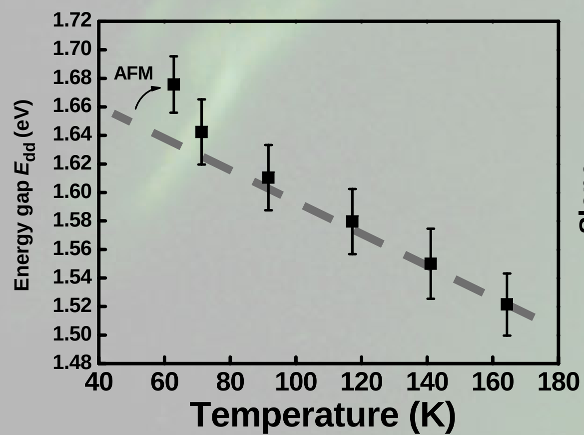
□ Observed the blueshift of energy gap !

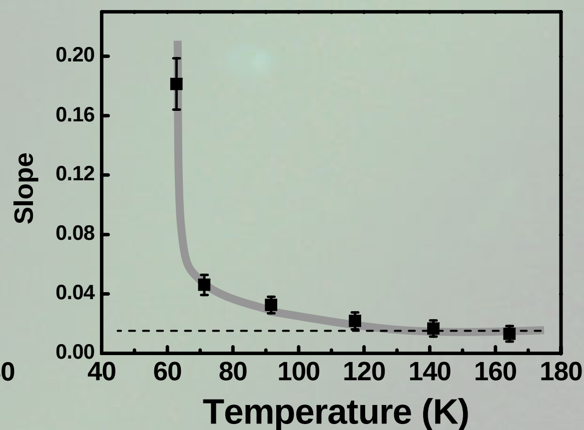
Ultrafast dynamics in $HoMnO_3$

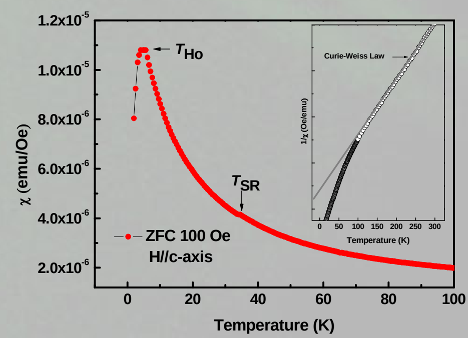
Charge transfer from e_{2g} to a_{1g} by pump pulses





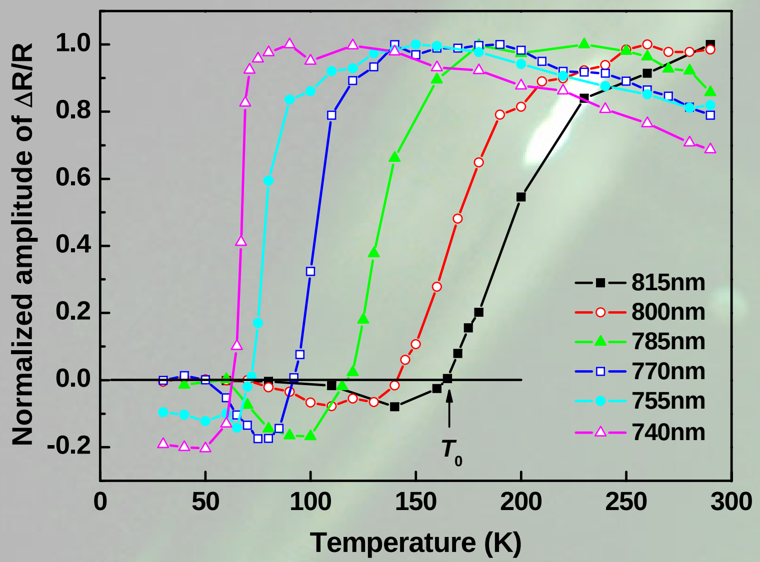


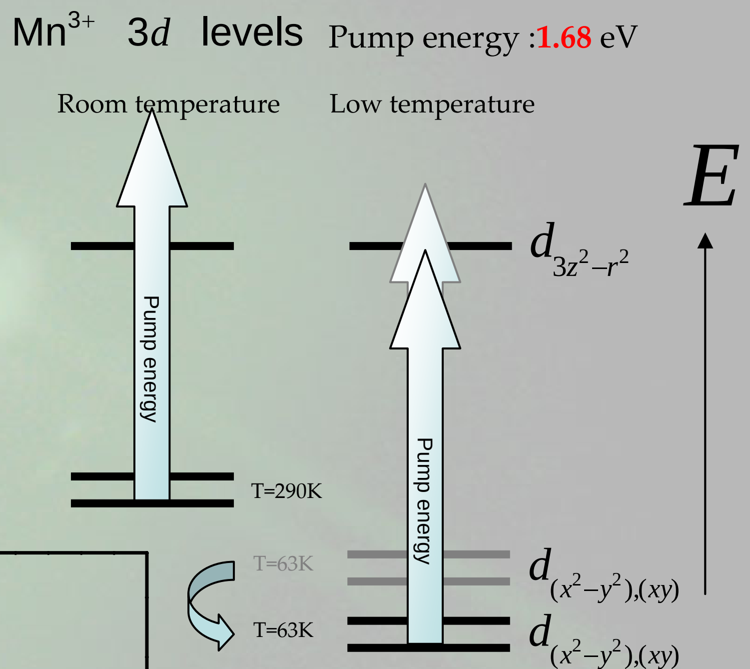


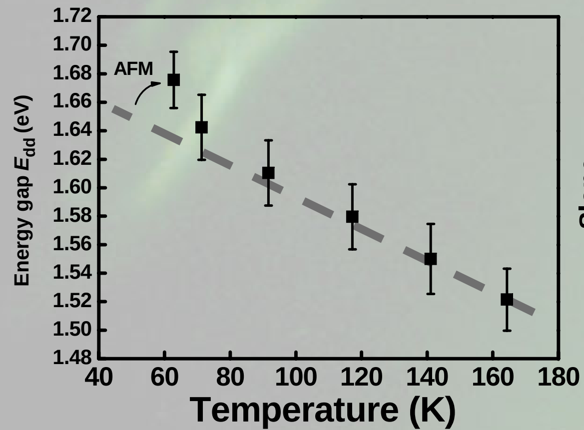


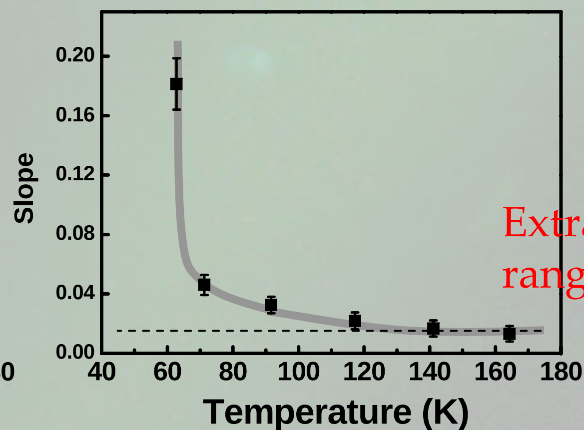
Ultrafast dynamics in $HoMnO_3$

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





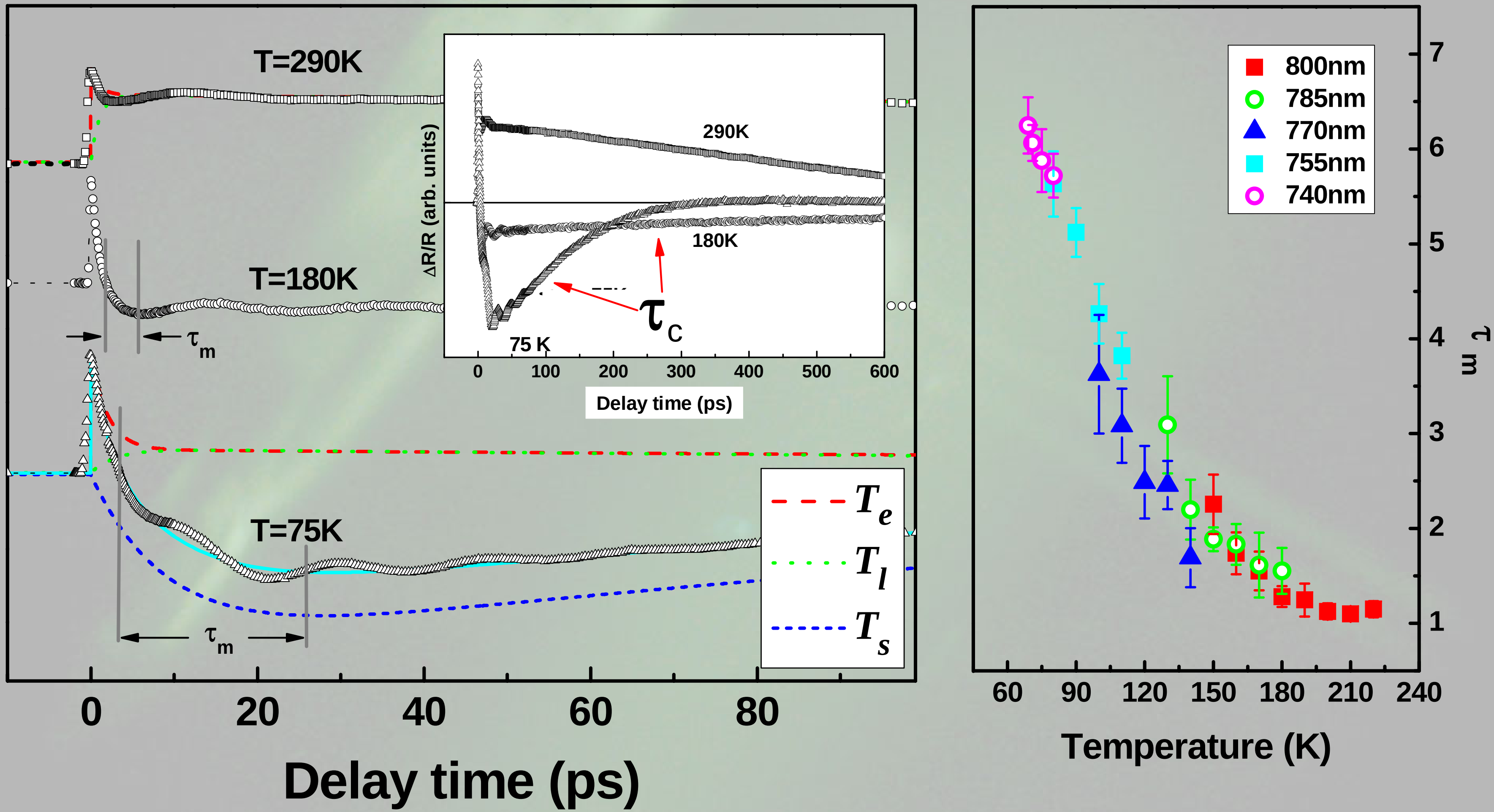




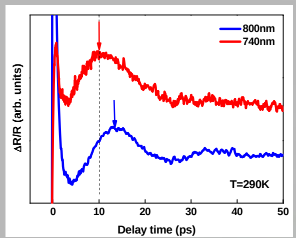
Extra-blueshift comes from long-range AFM ordering!!

Ultrafast dynamics in $HoMnO_3$

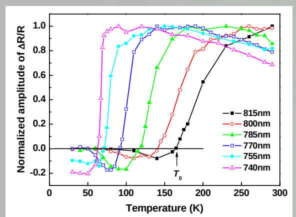
Demagnetization dynamics



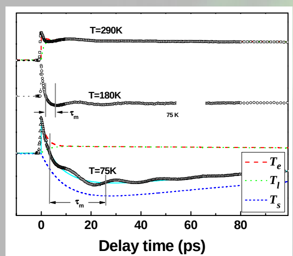
Summary



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



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

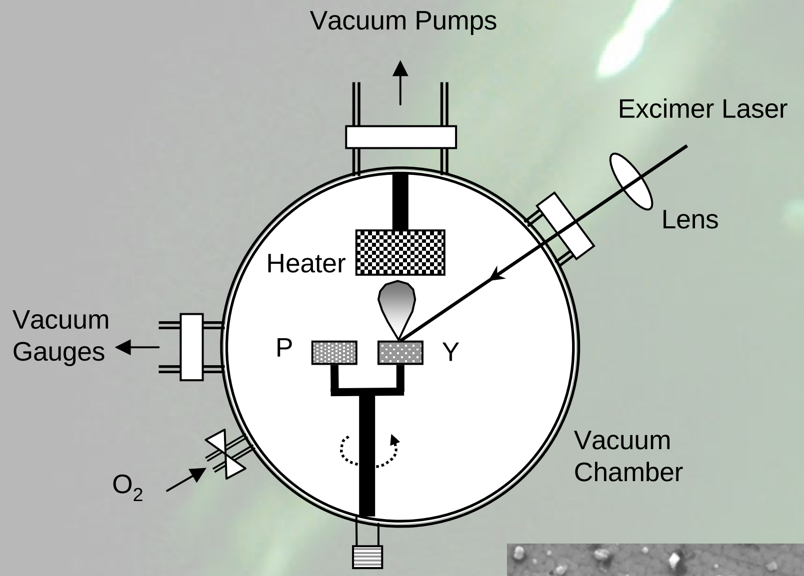


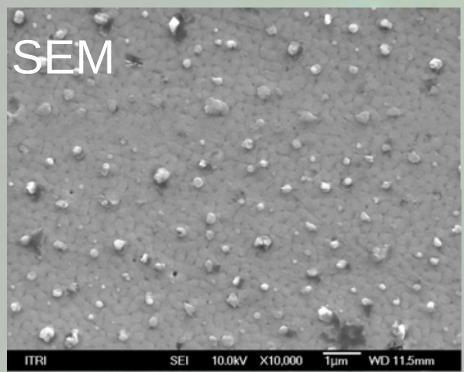
- The demagnetization time (τ_m) in a few ps scale and its recovering time (τ_c) in a few 100 ps scale were shown in the ΔR/R.

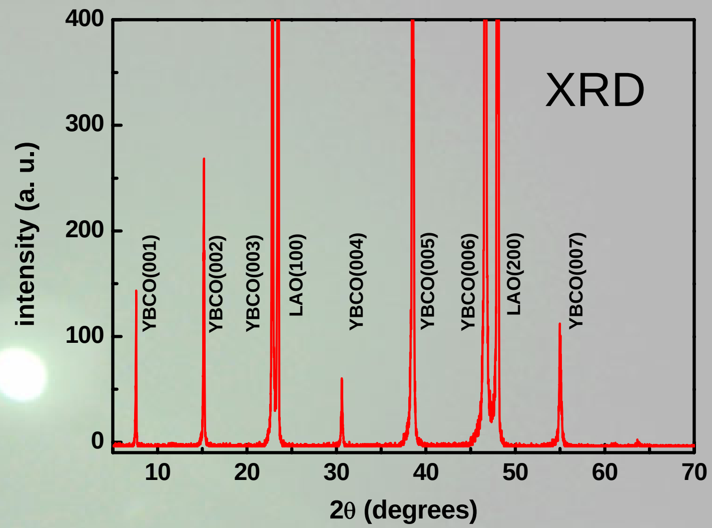
YBCO nanodots

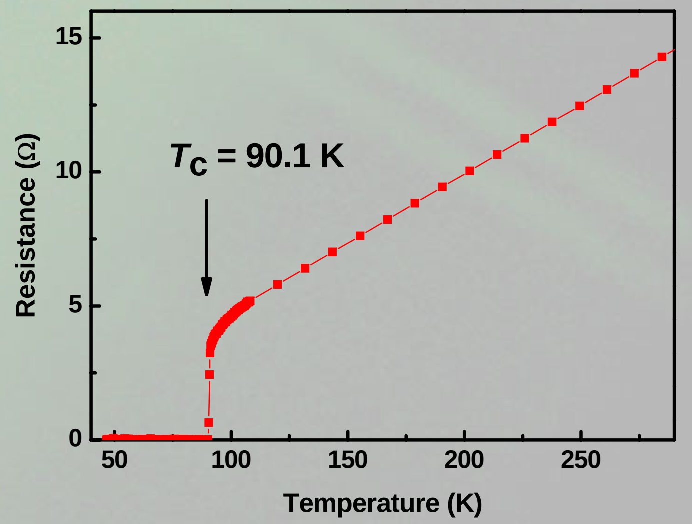
Sample reparation:

(001) $YBa_2Cu_3O_7$ (YBCO) / (100) $LaAlO_3$



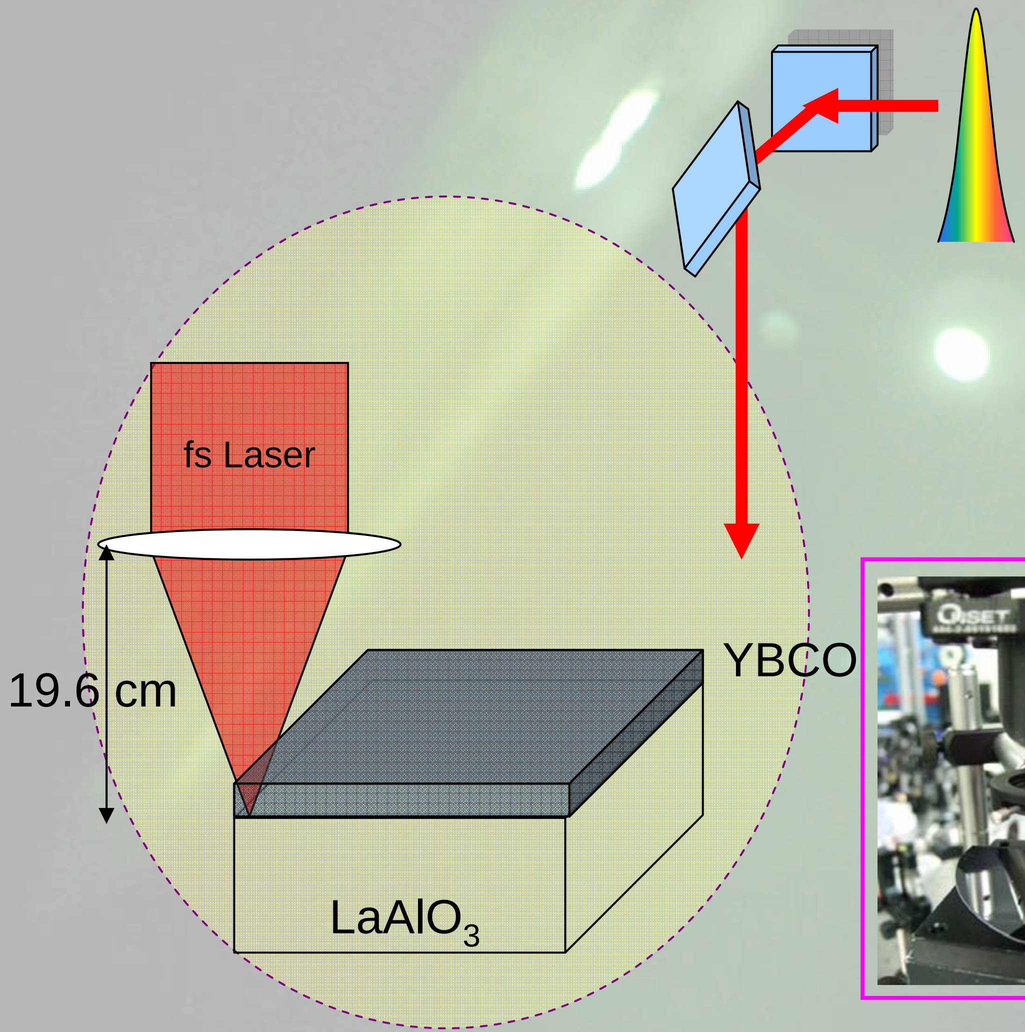


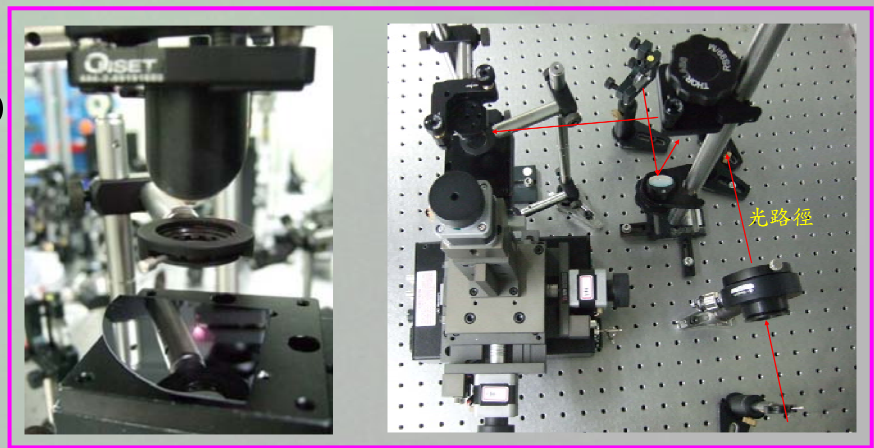




YBCO nanodots

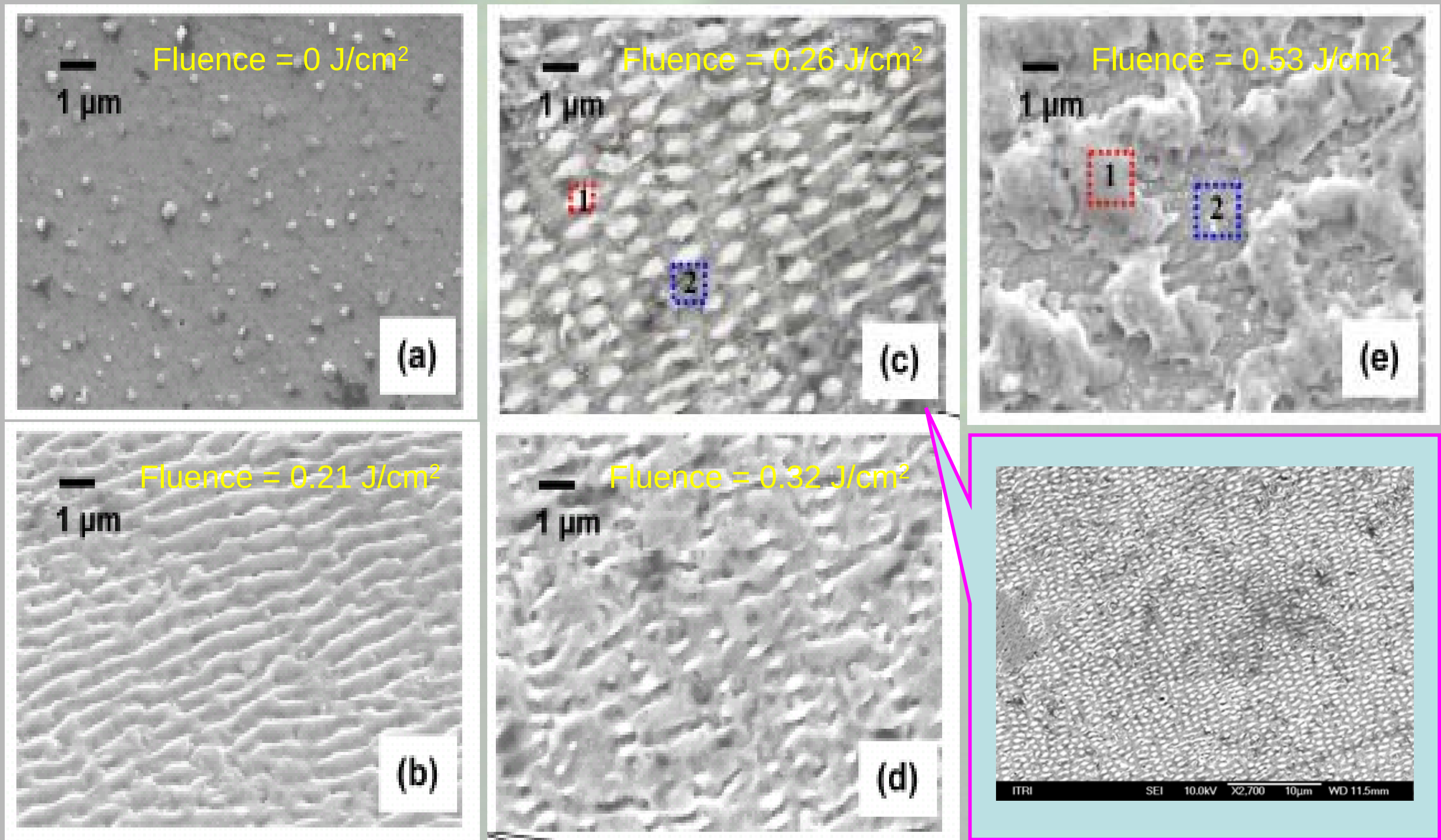
Experimental setup: (spot size~110 μm)





YBCO nanodots

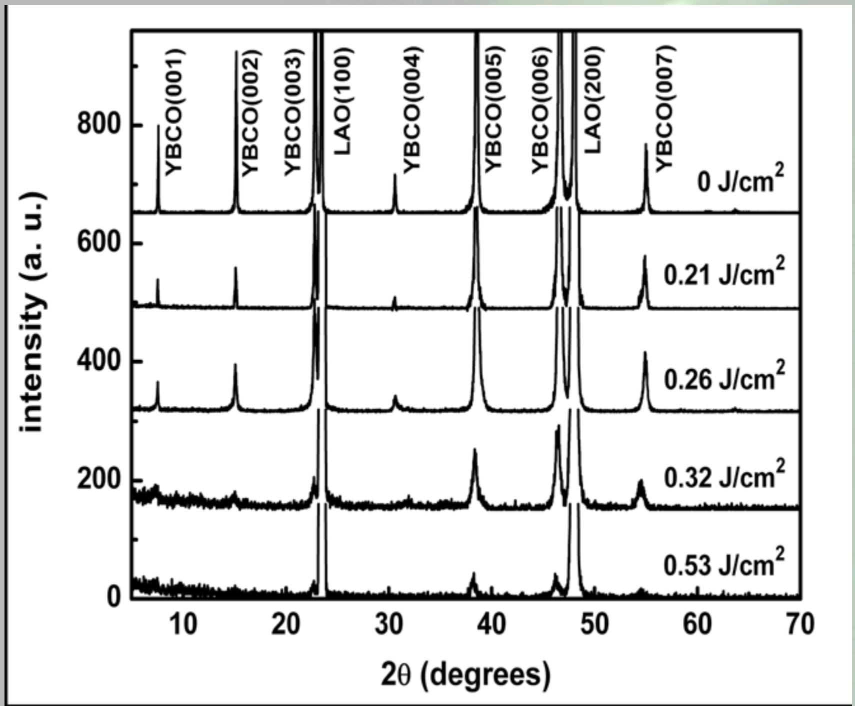
Results – surface morphology



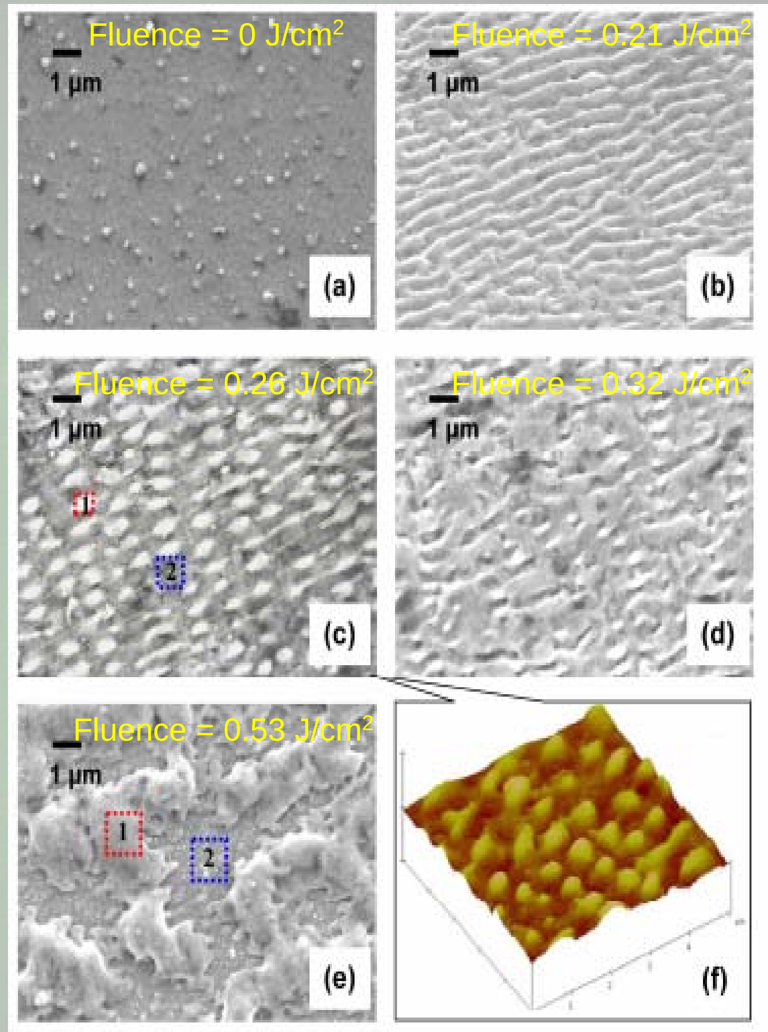
C. W. Luo, C. C. Lee, et al., Optics Express **16**, 20610 (2008)

YBCO nanodots

Results – structure

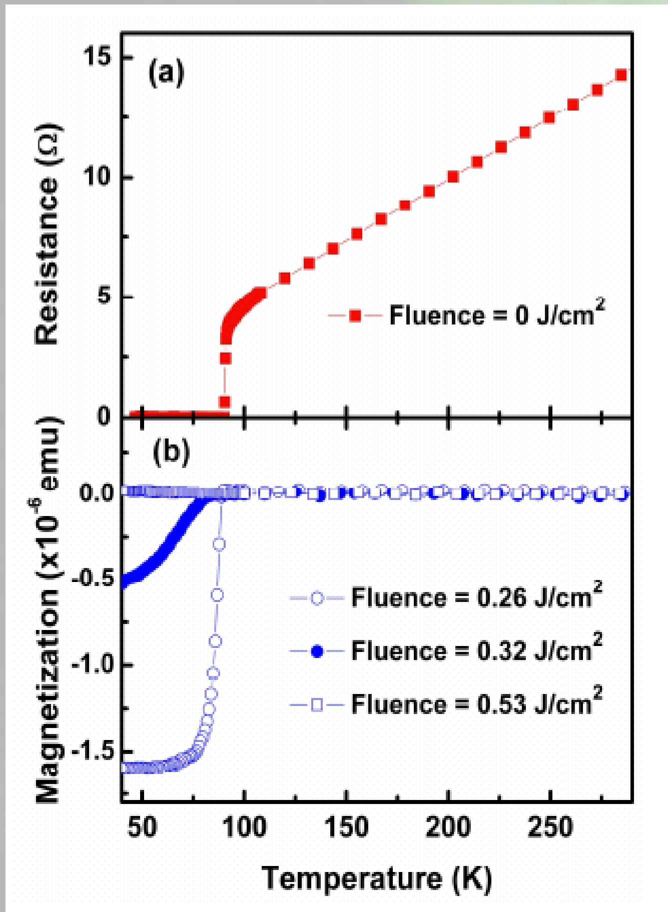


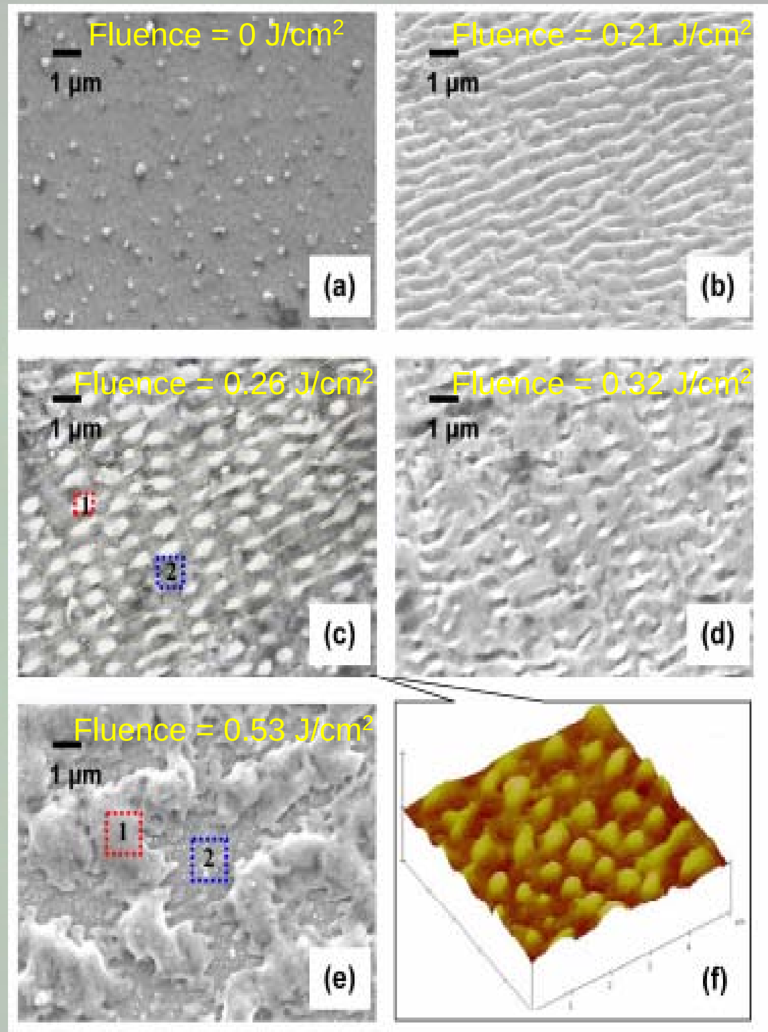
XRD signals of YBCO thin films at various laser fluences.



YBCO nanodots

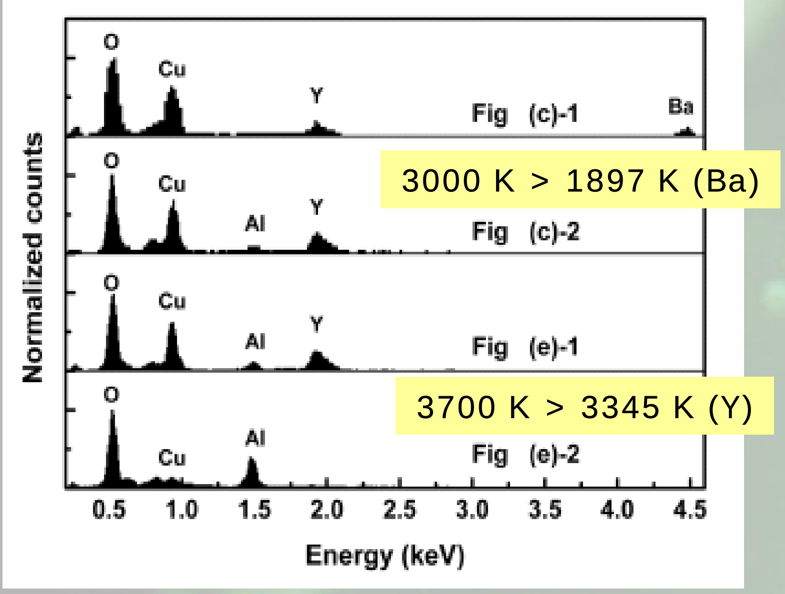
Results – superconductivity





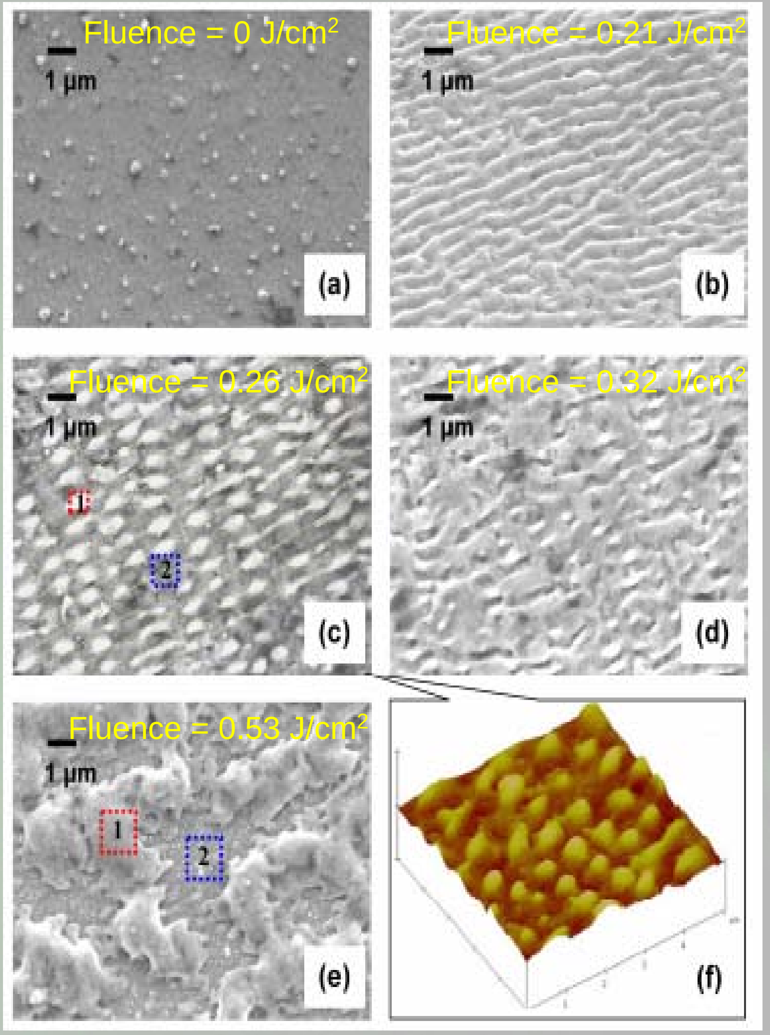
YBCO nanodots

Results – composition

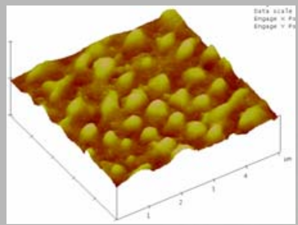


EDS spectra show the composition of area 1 and area 2.

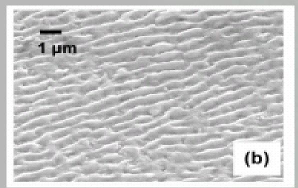
$$\Delta T = W/CV \quad \begin{cases} W \approx 0.1\,\mathrm{mJ} \\ C = 2.86\times10^{6}\ \mathrm{J\,m^{-3}K^{-1}} \\ V = 1.14\times10^{-14}\ \mathrm{m^{3}} \end{cases}$$



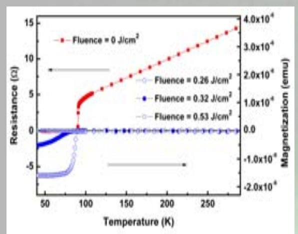
Summary



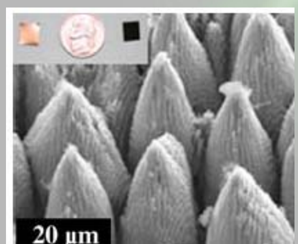
- The surface microstructure of YBCO thin films can be manipulated by properly controlling the fluence of the irradiating femtosecond laser.



- 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
B. K. Nayak, et al., Appl. Phys. A **90**, 399 (2008)

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Thank you

for your attention!!