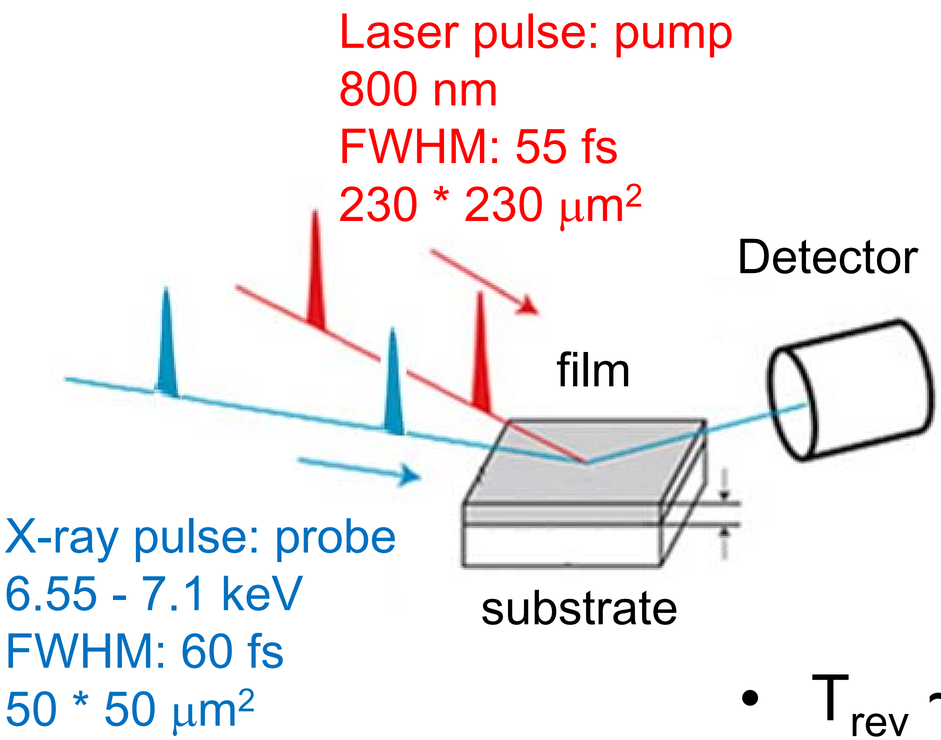
遷移金属酸化物の時間分解X線回折

H. Wadati[1,2], P. Beaud[3], U. Staub[3], T. Tsuyama[1,2],
N. Pontius[4], C. Schussler-Langeheine[4],
M. Nakamura[5], S. Chakraverty[5], H. Y. Hwang[5,6],
M. Kawasaki[1,5], and Y. Tokura[1,5]

[1]QPEC, Univ. of Tokyo, [2]ISSP Univ. of Tokyo, [3]PSI,
[4]HZB, [5]RIKEN CEMS, [6]Stanford Univ.

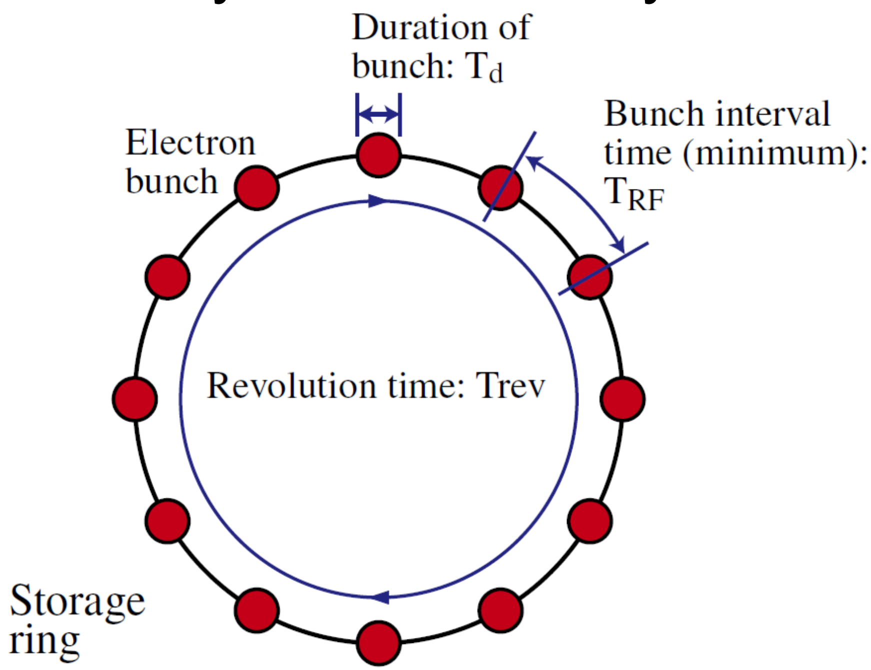
Time-resolved x-ray diffraction

<u>Experimental setup:</u>
Pump-probe



S. Yamamoto and I. Matsuda,
J. Phys. Soc. Jpn. **82**, 021003 (2013).

<u>Time structure</u>
of synchrotron x-ray



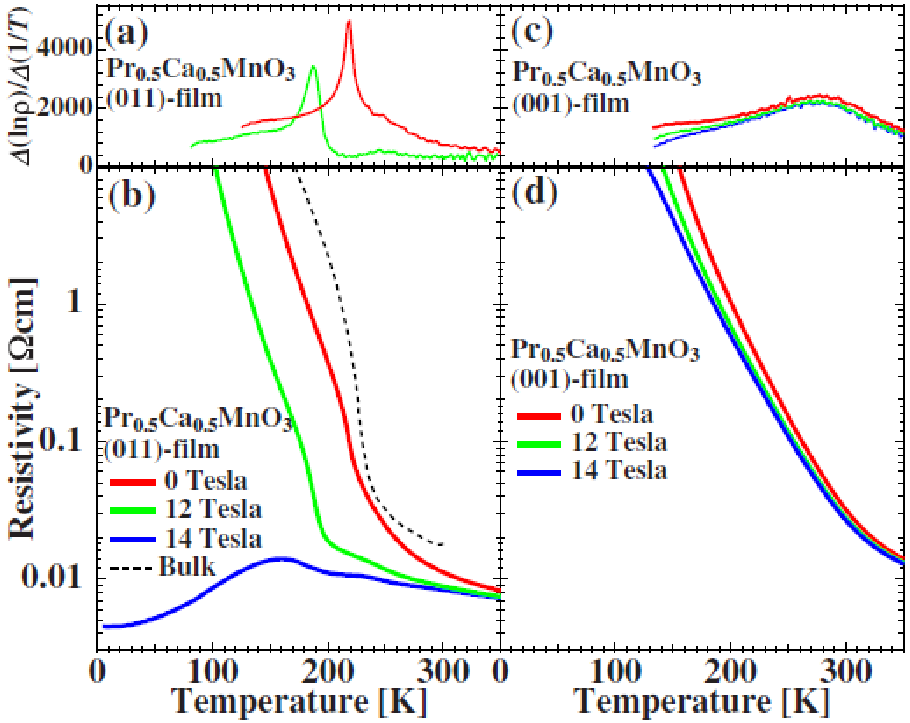
- T_{rev} ~ μs
- T_{RF} ~ ns
- T_d ~ ps

→ Δt ~ ps

To reach ~ fs, x-ray free electron laser (XFEL) is necessary!

Charge ordering in $Pr_{0.5}Ca_{0.5}MnO_3$ thin films

Electrical resistivity of $Pr_{0.5}Ca_{0.5}MnO_3$/LSAT



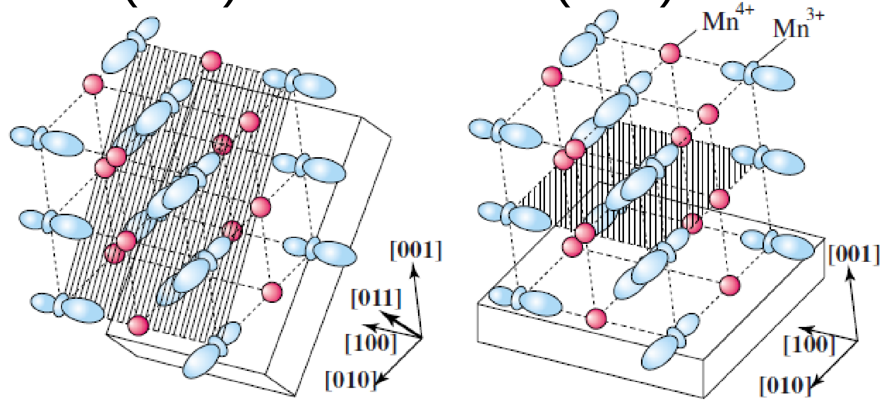
$T_{CO}^{(011)}$ ~ 220 K $T_{CO}^{(001)}$ ~ 300 K

T_{CO}^{bulk} ~ 230 K

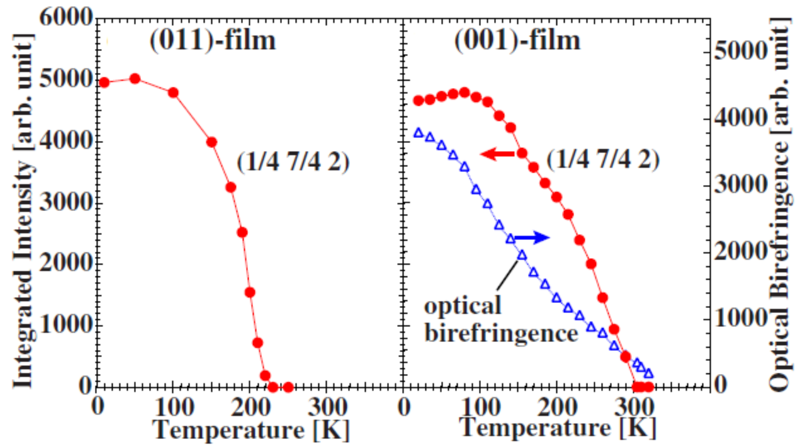
LSAT:
$(LaAlO_3)_{0.3}$-$(SrAl_{0.5}Ta_{0.5}O_3)_{0.7}$

Charge and orbital ordered states

(011)-film (001)-film



Intensity of (1/4, 1/4, 0)



D. Okuyama *et al*., APL **95**, 152502 (2009).

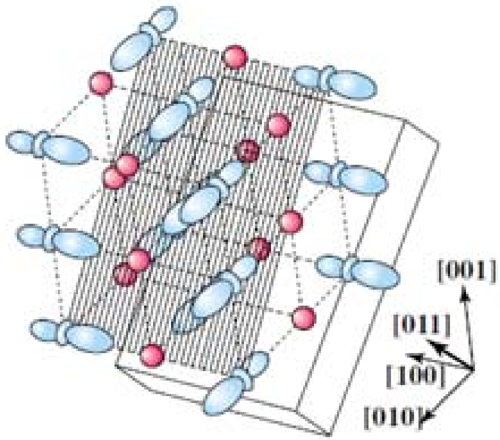
Samples

Thin films fabricated by pulsed laser deposition.

- $Pr_{0.5}Ca_{0.5}MnO_3$ (~ 50 nm)/LSAT(011)

growth conditions

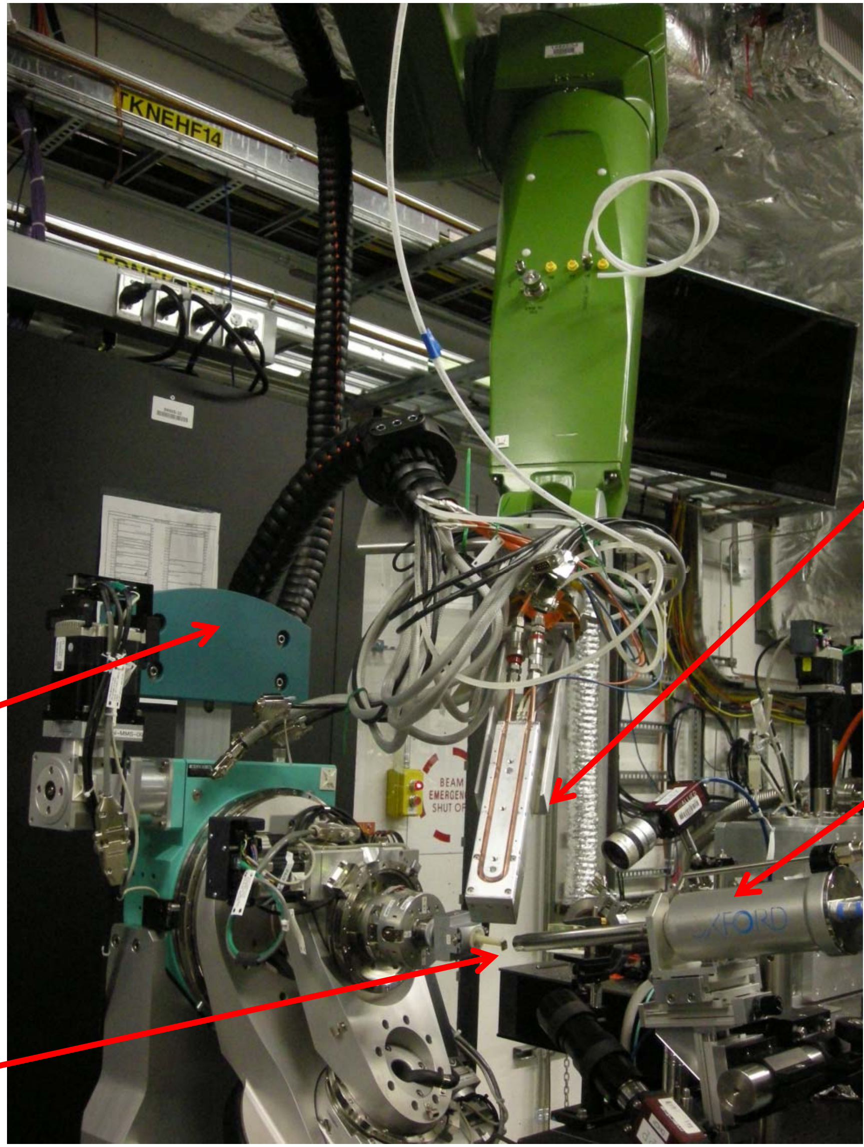
T_{sub} = 850 °C, P_{O_2} = 1.5 mTorr



Measurements

- LCLS (Linac Coherent Light Source)
- XPP (X-ray pump-probe)
- 150 K (Liquid N_2)
- $h\nu$ ~ 6.5 keV (near Mn K edge)

Experimental setup



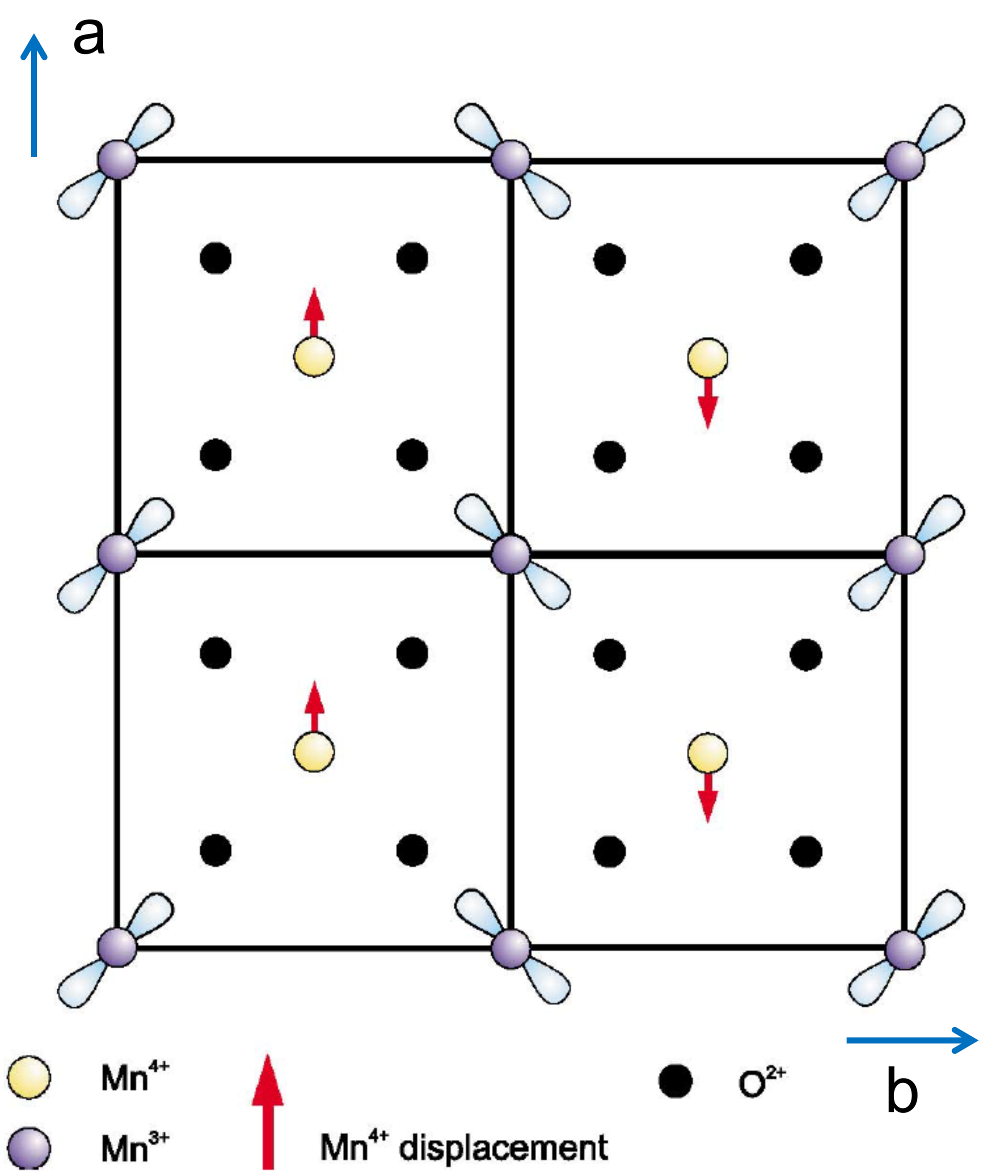
Detector
CS-PAD 140K

Liquid N_2

Goniometer

Thin film

Ordering peaks



From structure
(2 1/2 0) peak
Mn^{4+} displacement

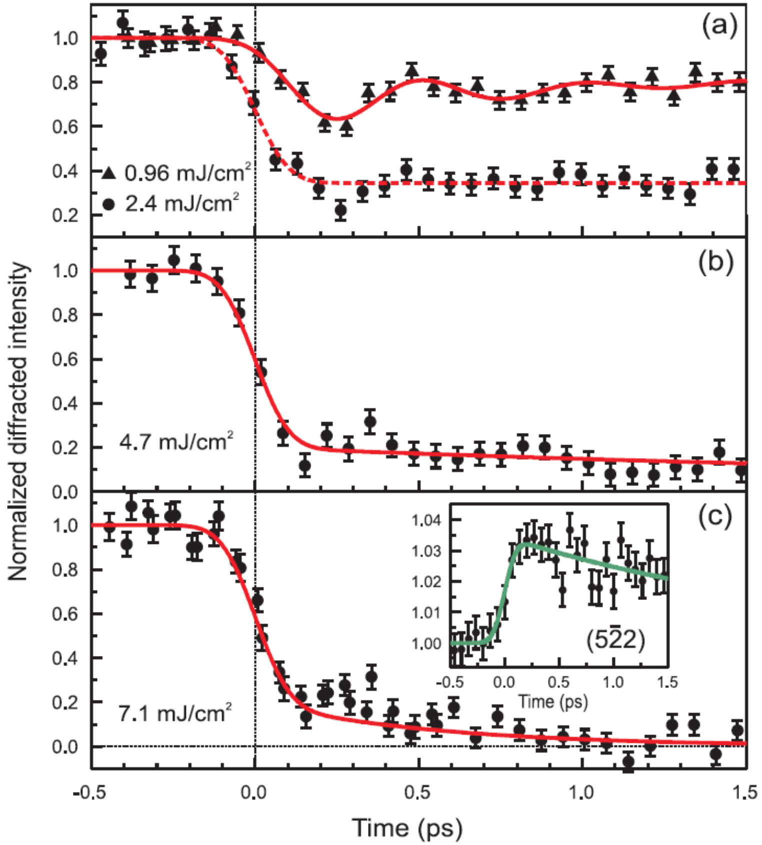
From orbital ordering
(0 5/2 0) peak

From charge ordering
(0 3 0) peak

S. B.Wilkins *et al.*, Phys. Rev. Lett. **91**, 167205 (2003).

$La_{0.42}Ca_{0.58}MnO_3$/MgO(001) film

(5 5/2 2) peak
(mainly from structure)
7.15 keV (off) at SLS



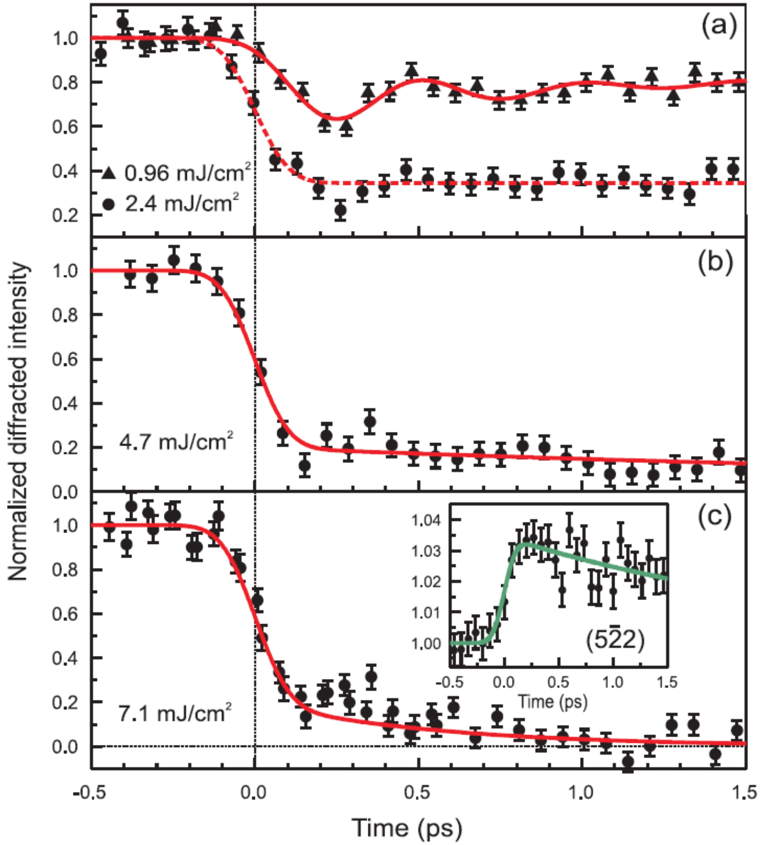
$$I(t)/I(0) = 1 - Ae^{-t/\tau_1}(1 - e^{-t/\tau_2}\cos 2\pi\nu t)$$

ν = 1.98 THz (0.5 ps)
~ 70 cm^{-1}
(phonon: A_g mode)

P. Beaud *et al.*, PRL **103**, 177502 (2009).

$La_{0.42}Ca_{0.58}MnO_3$/MgO(001) film

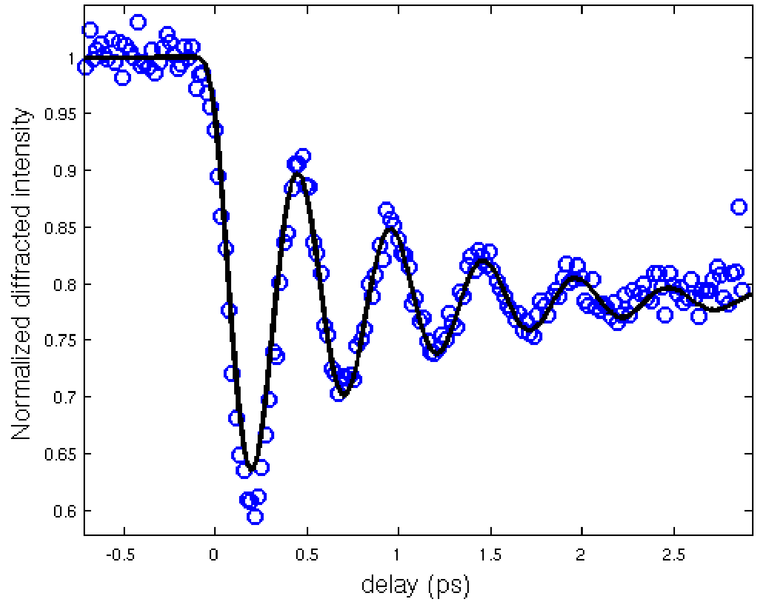
(5 5/2 2) peak
(mainly from structure)
7.15 keV (off) at SLS



P. Beaud *et al.*, PRL **103**, 177502 (2009).

(5 5/2 2) peak

7.09 keV (off) at LCLS



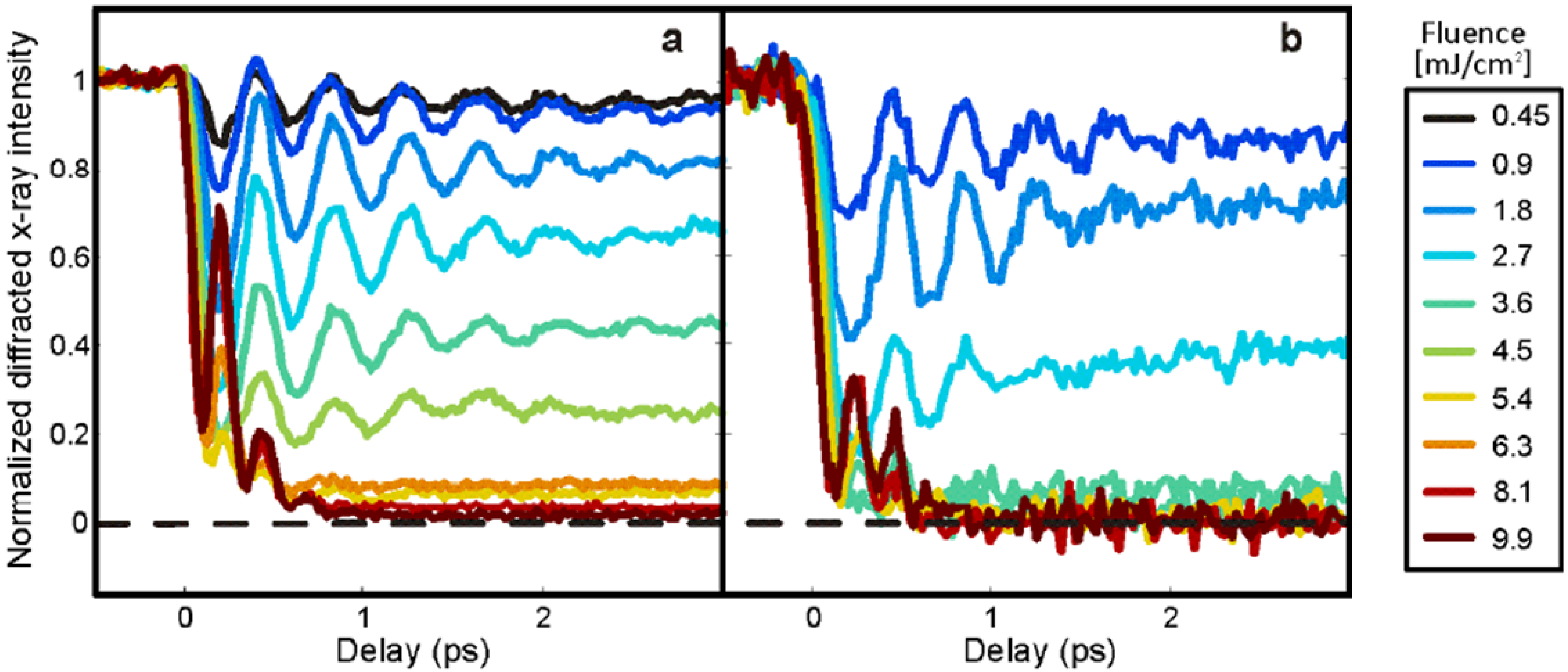
ν = 1.98 THz (0.5 ps)

Laser fluence: 2 μJ

$Pr_{0.5}Ca_{0.5}MnO_3$/LSAT(011) (exp.)

(2 1/2 0) peak
(mainly from structure)
6.53 keV (off)

(0 5/2 0) peak
(orbital ordering)
6.553 keV (on)

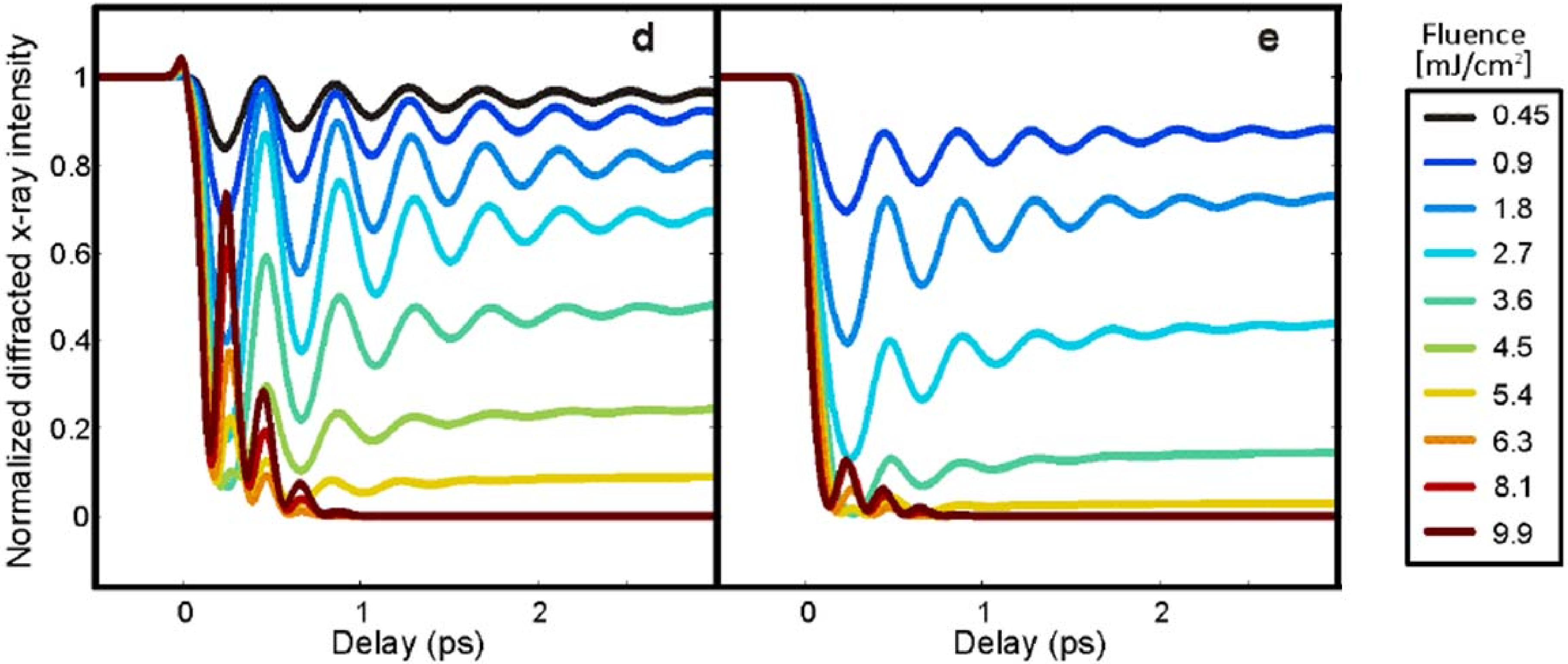


Oscillations due to coherent phonon (~ 2.5 THz).
Frequency doubling at higher fluence.

$Pr_{0.5}Ca_{0.5}MnO_3$/LSAT(011) (theory)

(2 1/2 0) peak
(mainly from structure)
6.53 keV (off)

(0 5/2 0) peak
(orbital ordering)
6.553 keV (on)

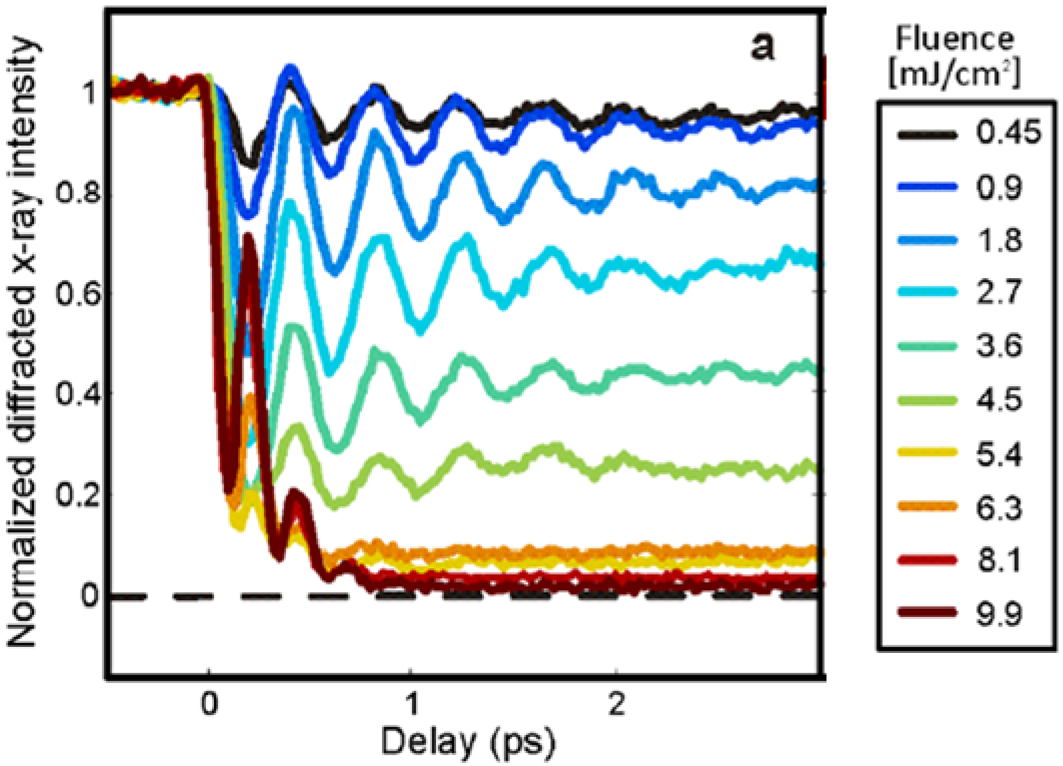


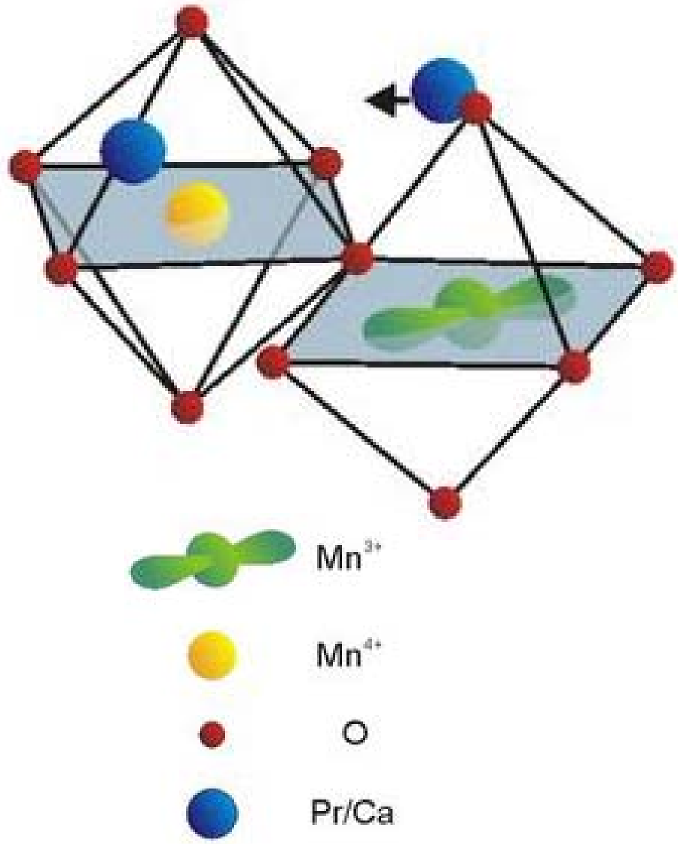
$$V(t) = -\frac{a}{2}\eta_t^2 y_1^2 + \frac{b}{4} y_1^4 + \frac{c_{21}}{2}(y_2 - y_1)^2 + \frac{c_{32}}{2}(y_3 - y_2)^2 + \frac{c_{43}}{2}(y_4 - y_3)^2$$

Jahn-Teller mode & breathing mode   Slower coherent atomic motions

Summary 1

Melting orbital ordering



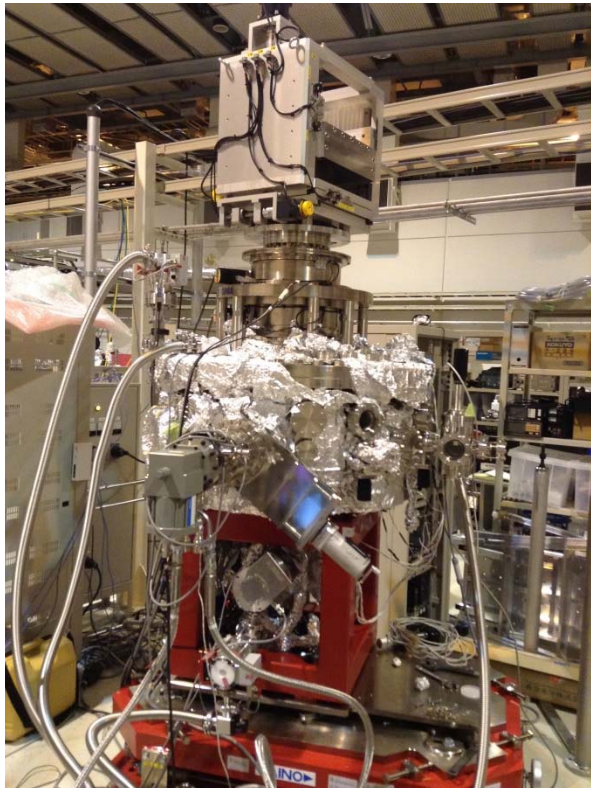


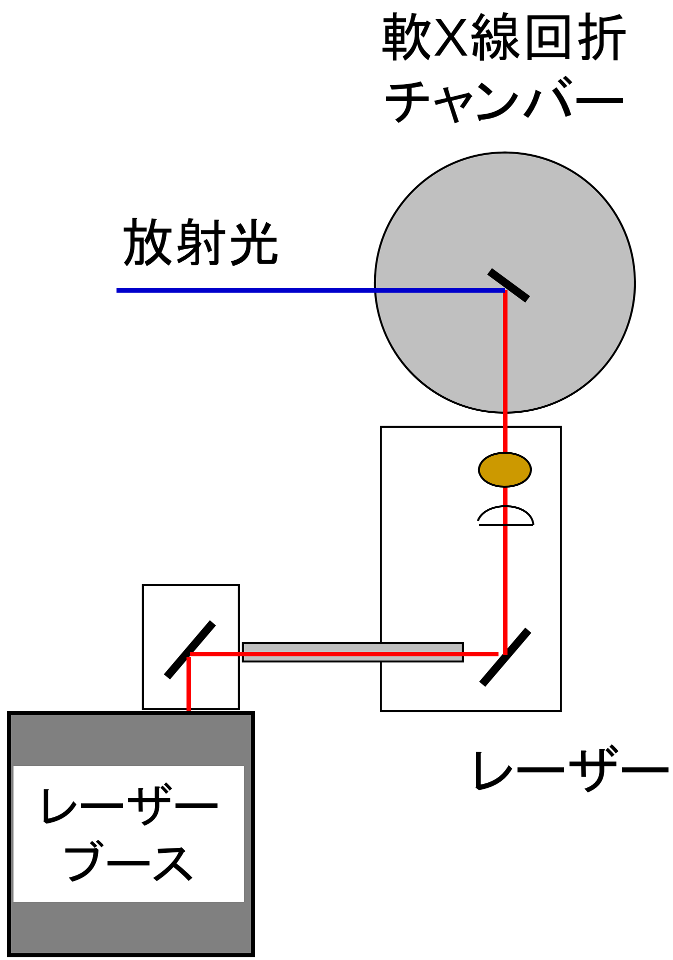
Coherent phonon of ~ 2.5 THz: Motion of Pr/Ca
Frequency doubling at higher fluence.

P. Beaud, H. Wadati *et al.*, Nature Materials **13**, 923 (2014).

東大物性研BL07LSUでの取り組み

軟X線回折チャンバー





東大物性研ビームライン
BL07LSUで今年2月から稼働