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Title:	Ultrafast all-optical manipulation of interfacial magnetoelectric coupling
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Ultrafast all-optical manipulation of interfacial magnetoelectric coupling

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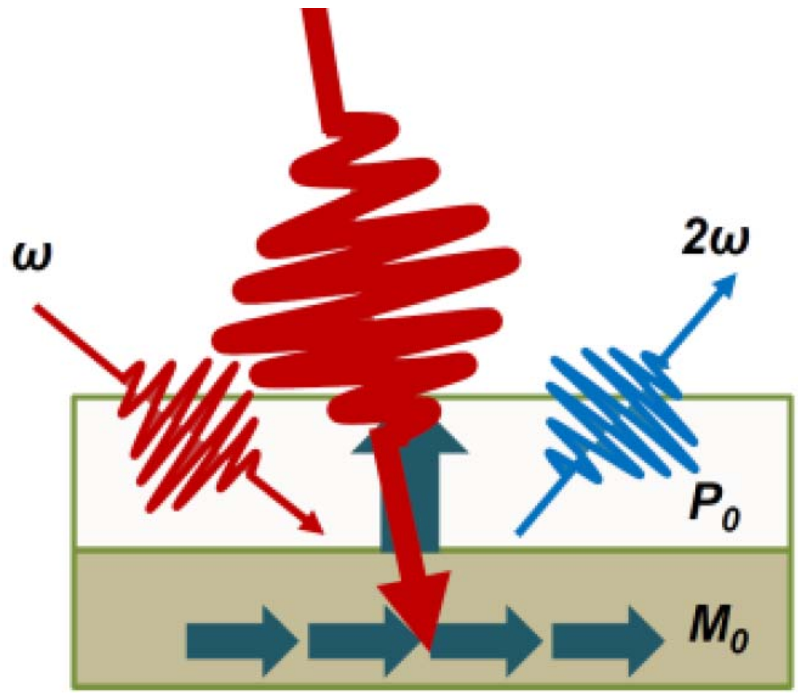


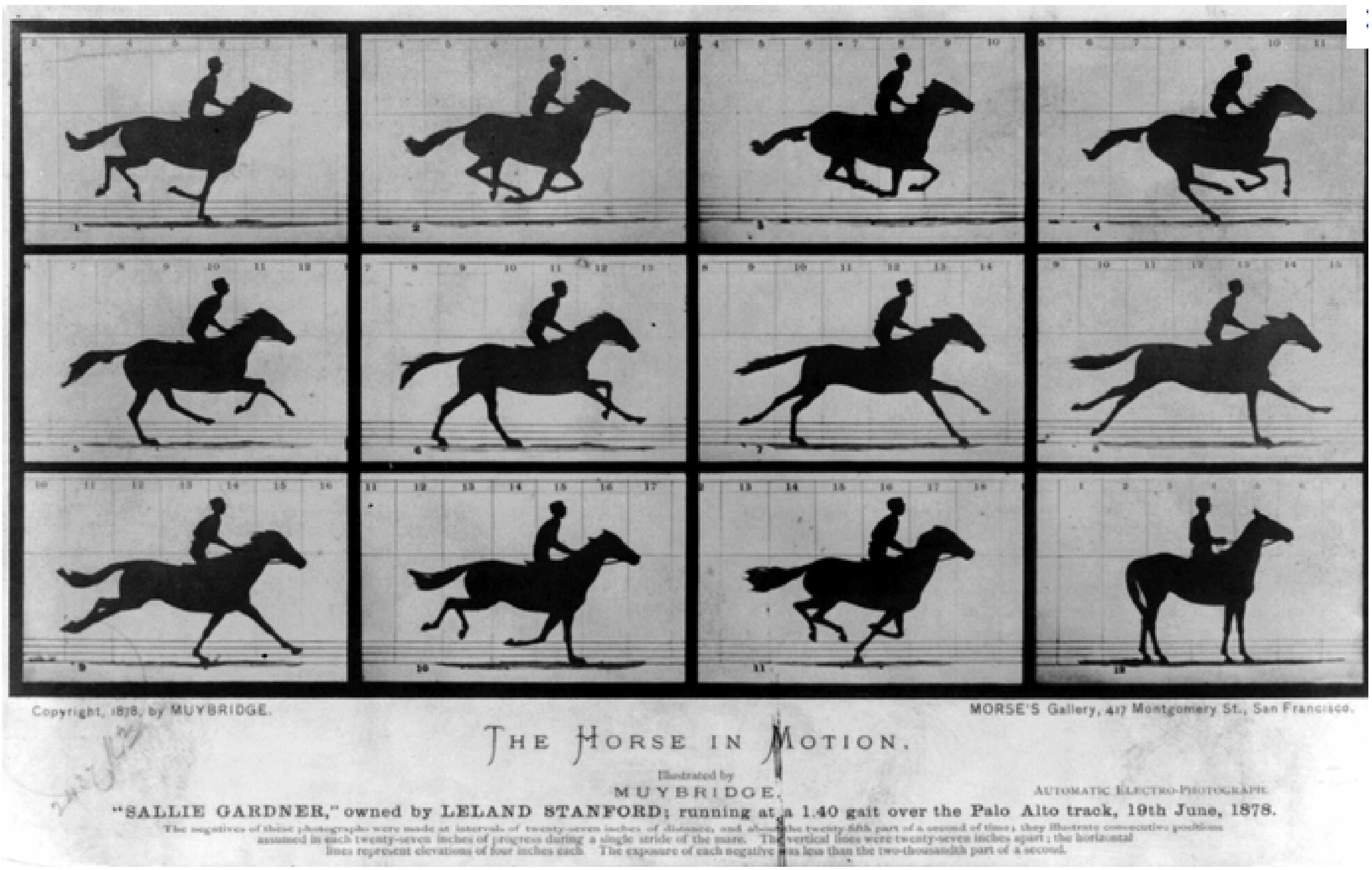
Outline

- **Introduction: ultrafast science , multiferroics and interfacial coupling**

1. **What is ultra fast science**
2. **Ferroelectric (FE) and piezoelectric effect**
3. **Ferromagnetism (FM) and magnetostriction**

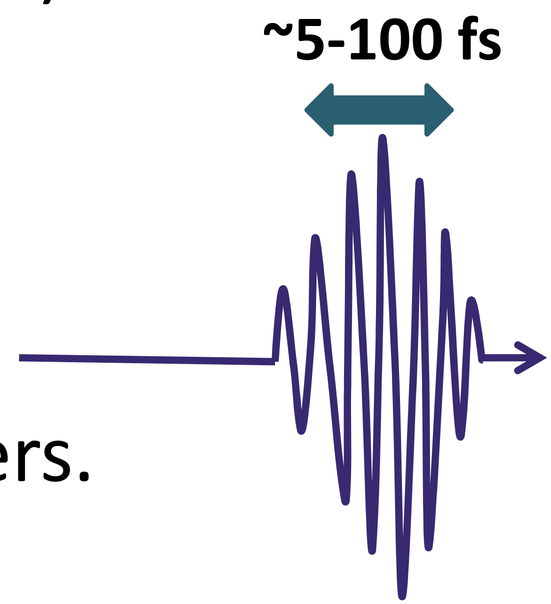
- **Optical second harmonic generation (SHG) , FE versus strain of $Ba_{0.1}Sr_{0.9}TiO_3$ (BSTO)**
- **Quasiparticle dynamics in FM $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) upon photoexcitation**
- **How to couple FE and FM orders at an interface by ultrafast optical methods**
- **Experimental evidences of optically induced ME coupling**
- **Conclusion**

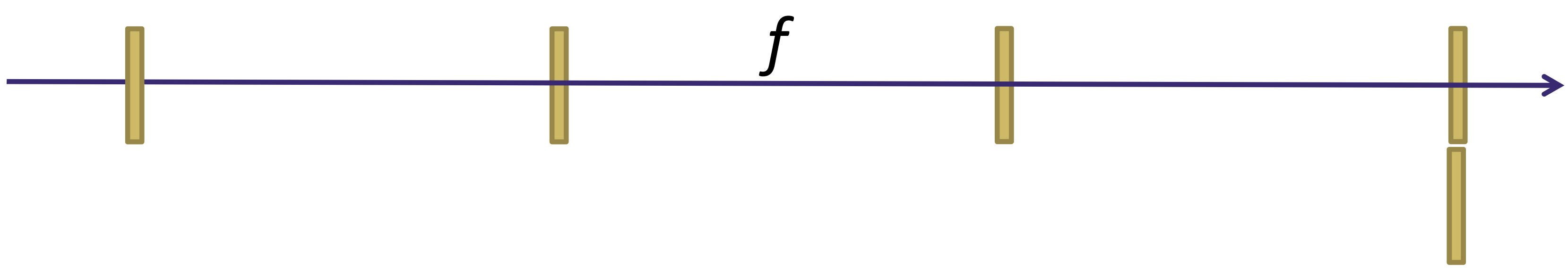
The first movie, 1878



What is ultrafast science?

- Science, physics properties, chemical reactions on time scale of 10^{-9}-10^{-15} s (ns-fs) or faster.
- Detectors are slow.
- Science movie.
- We need **timing** and **snapshot**.
- Pulses, bandwidth, ultrafast lasers.

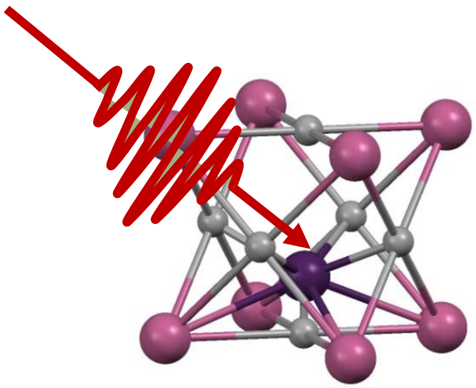




Generation of light sources

Optical frequency mixing

Spectroscopy



Second harmonic generation

Surface probe, domain imaging
Crystal symmetry studies

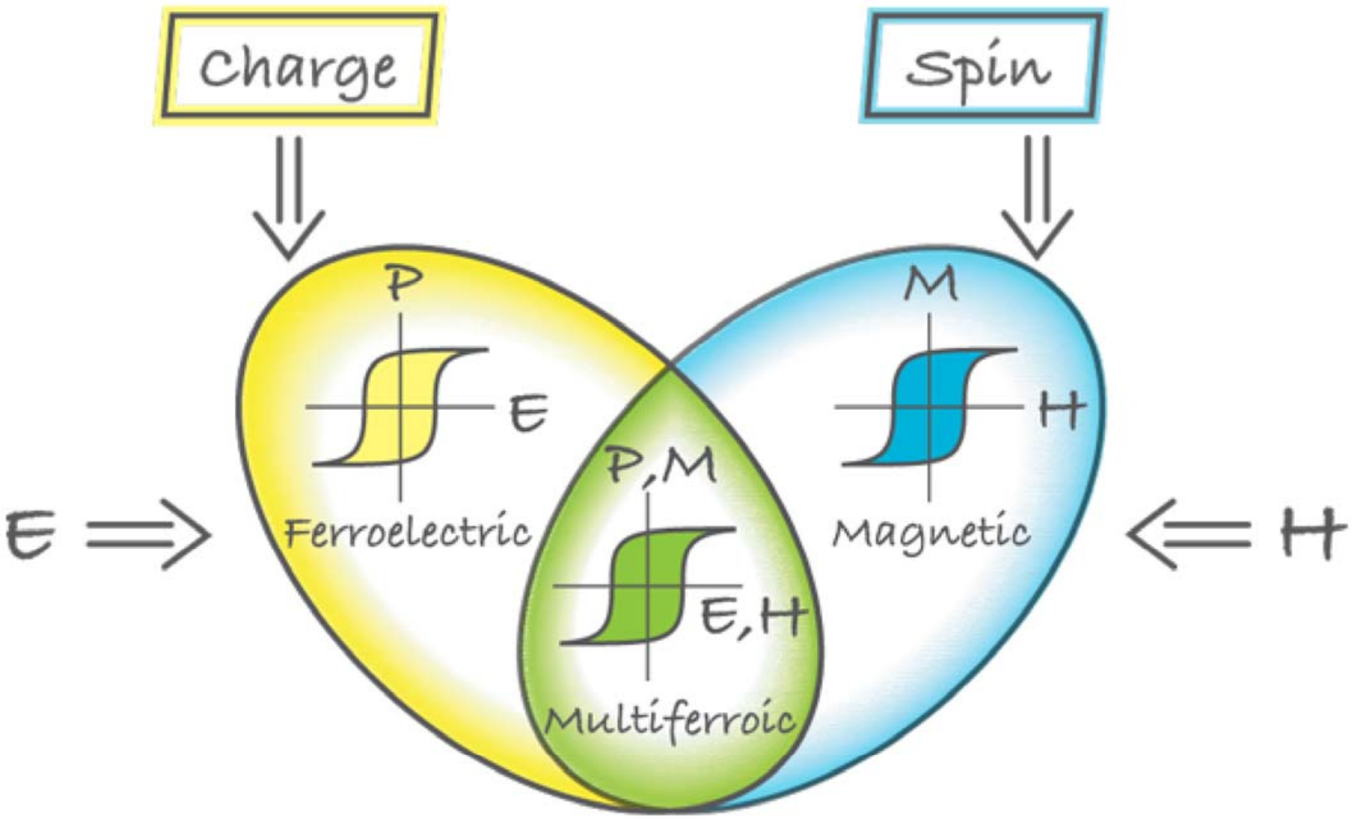
High harmonic generation
X-UV, x rays…

Element sensitive probes,
Imaging, diffractions

THz generation

Quasi-particle excitation and
detection, meta-materials

Magnetoelectric Mutiferroics



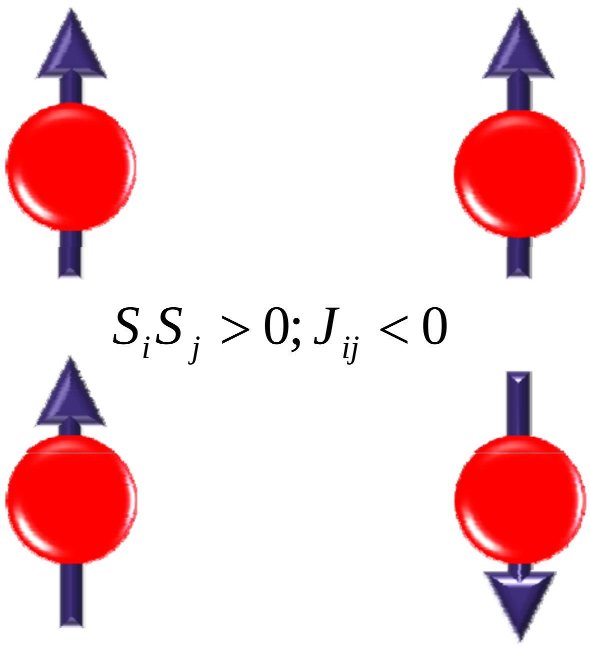
- **Hot topics in physics and material science: how to enhance coupling**
- **E-field control of M: data storage applications**

Coupling is still weak in single phase materials !!

Cartoon courtesy of Khomskii , Physics 2, 20 (2009)

Elastic coupling

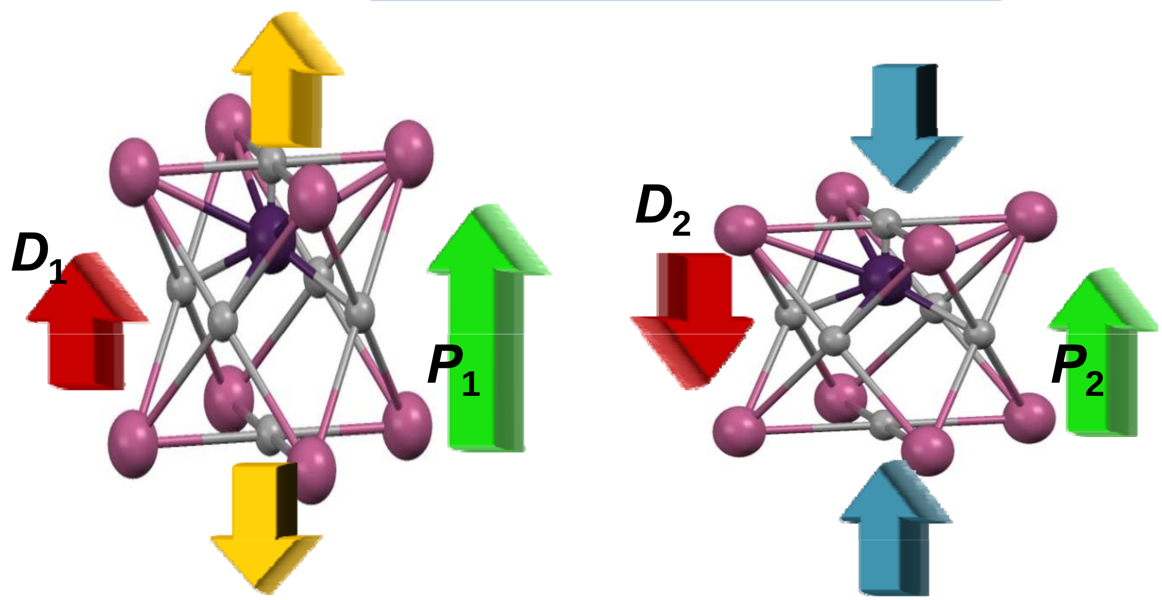
Magnetostriction



$$S_i S_j > 0; J_{ij} < 0$$

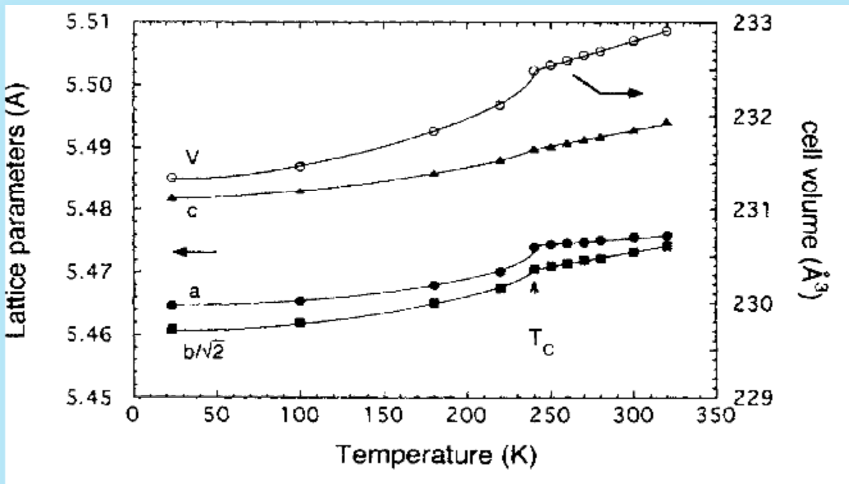
$$S_i S_j < 0; J_{ij} > 0$$

Piezoelectric effect

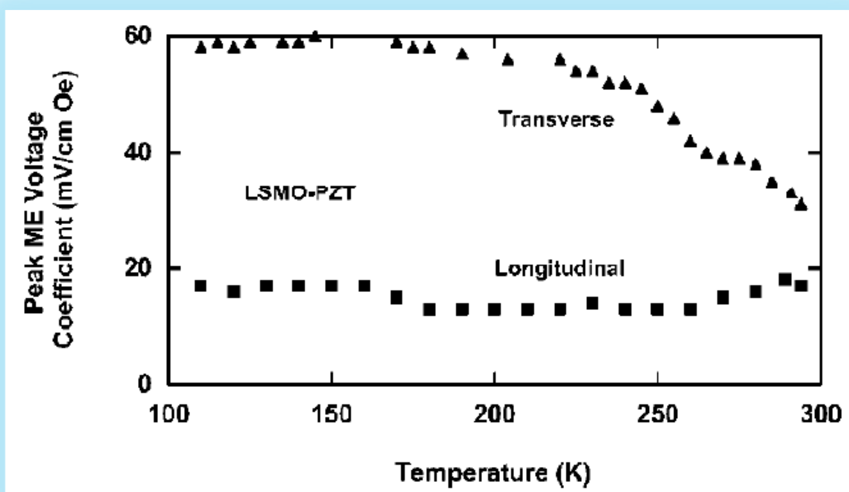


Indirect coupling through lattice/elastic properties is promising!

Magnetostric and piezoelectric examples



P. G. Radaelli, *et al.*, Phys. Rev. Lett. 75, 4488 (1995).

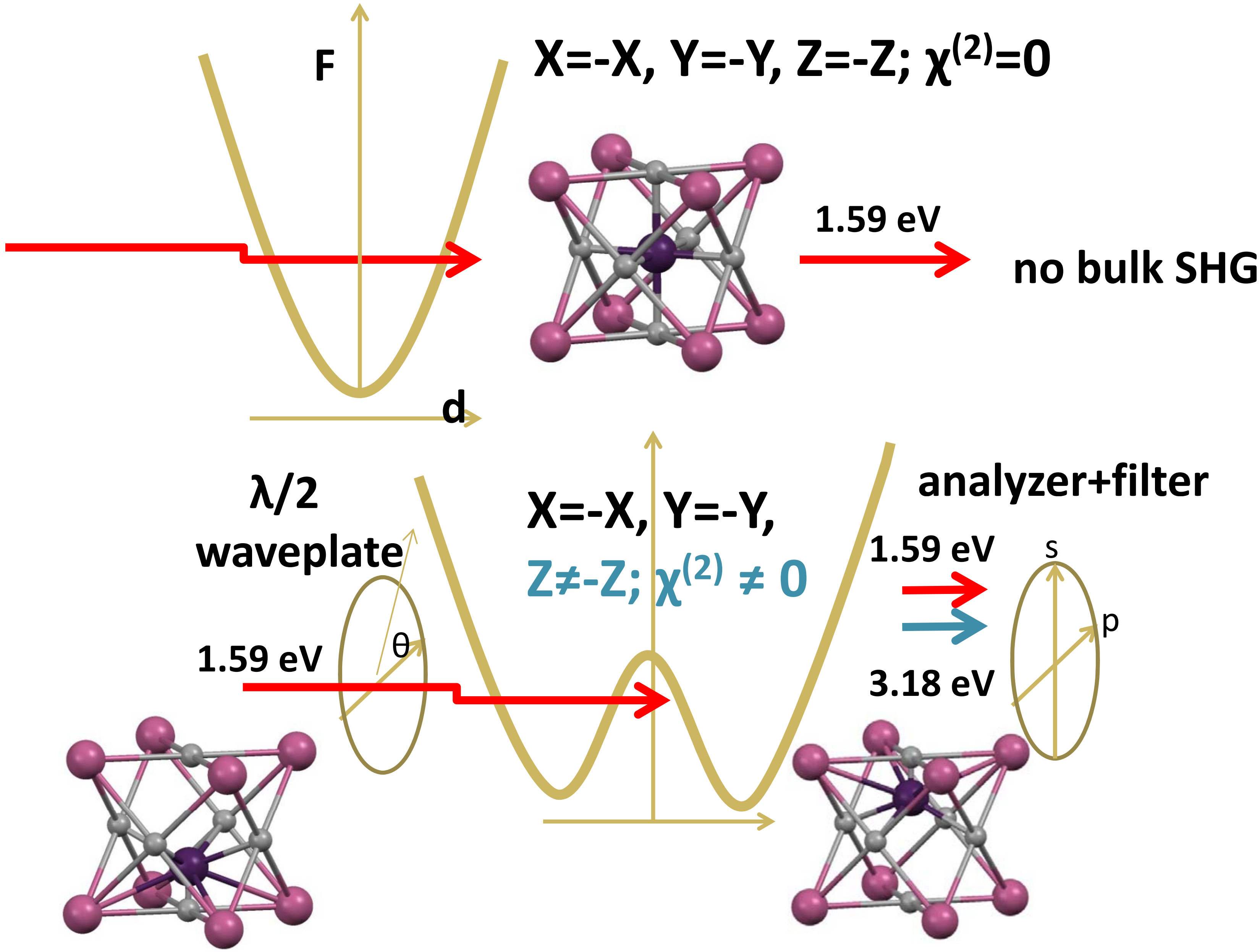


G. Srinivasan, *et al.*, Phys. Rev. B 65, 134402 (2002).

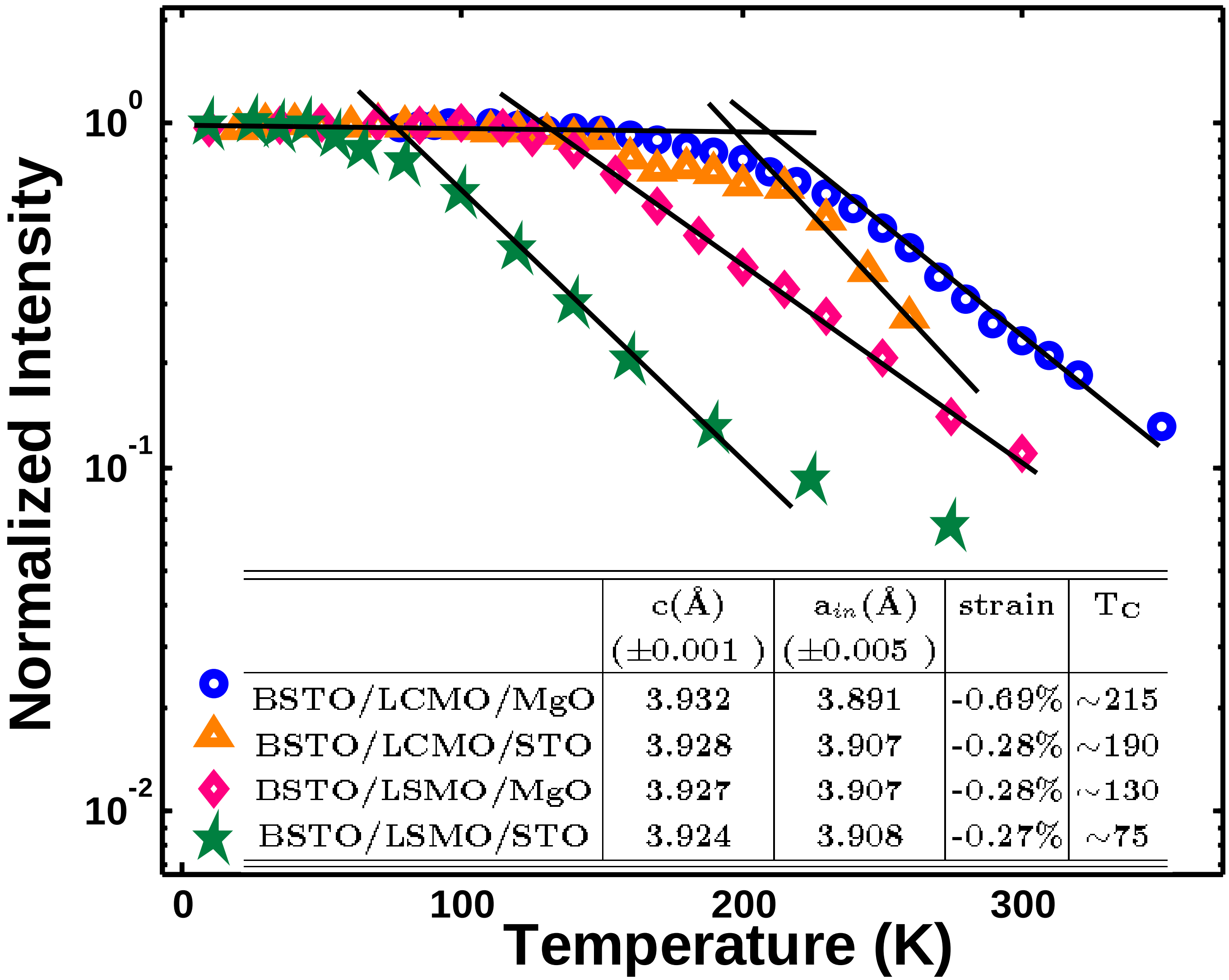
- Upon cooling across T_c, the lattice of LCMO contracts further than the regular contraction, due to ordered spins.
- Static ME coupling has been achieved in a FM/FE heterostructure of LSMO/PZT via applying an external B field.
- The lattice contraction of LSMO due to ordered spins causes a piezoelectric response along the sample normal (normal to interface).
- The largest ME effect occurs below the FM $T_{c.}$

LC(S)MOs are good candidates of magnetostrictive materials.

FEs are good piezoelectric materials



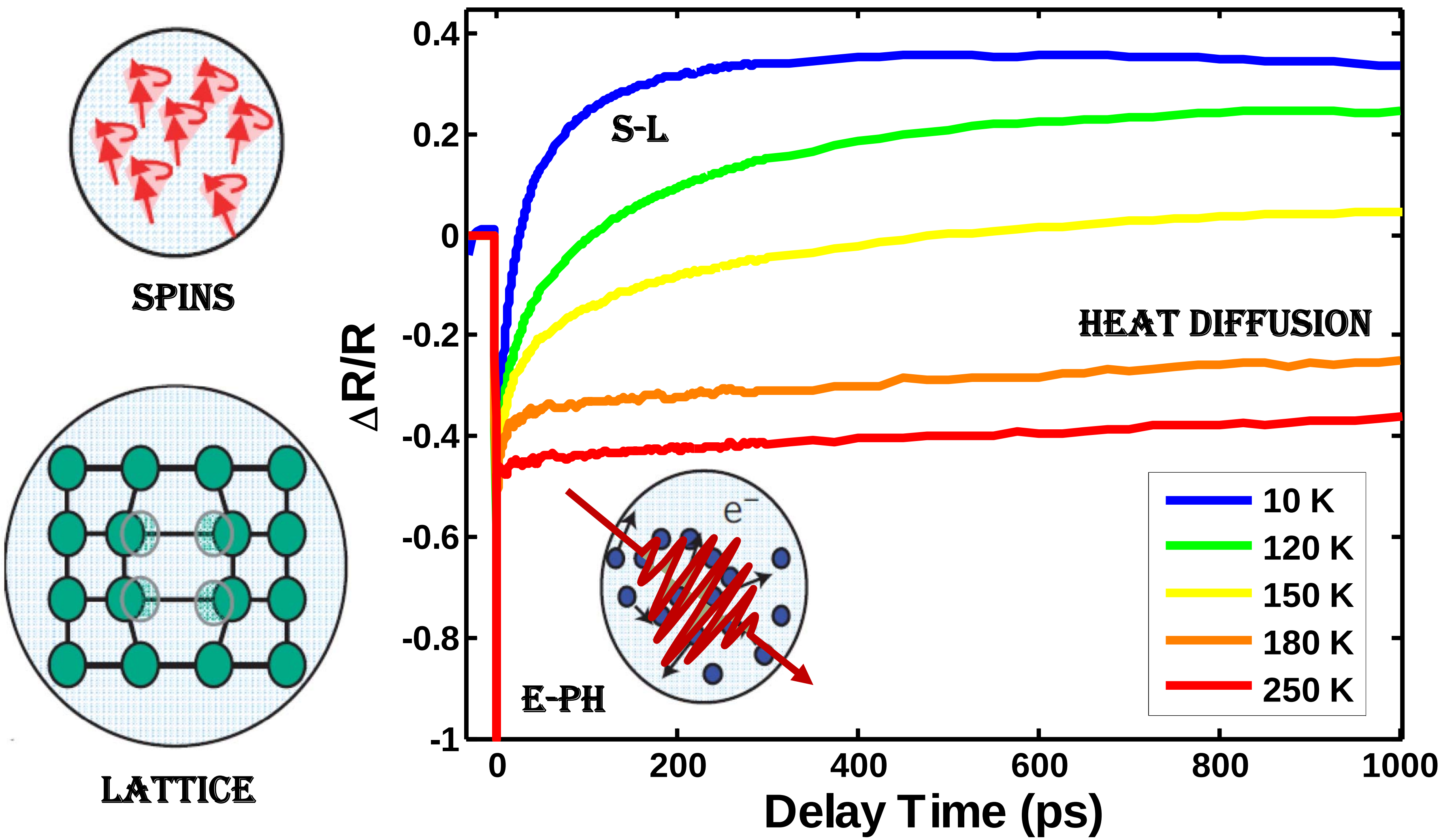
BSTO (FE) and strain



BSTO is an ideal piezoelectric material !

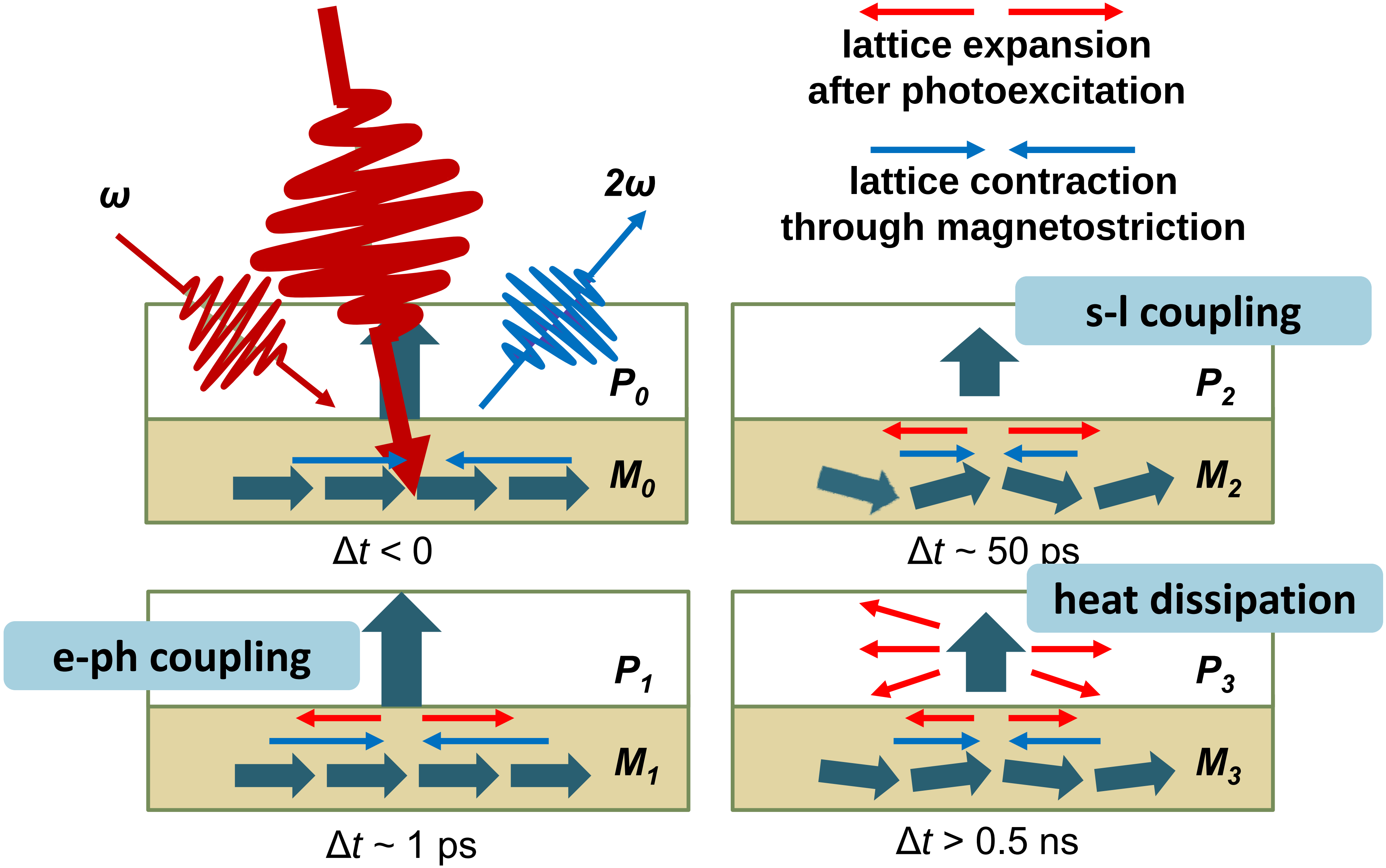
Sheu, *et al.* , PRB Rapid Comm. **88**, 202101(R) (2013)

Quasiparticle dynamics of LCMO

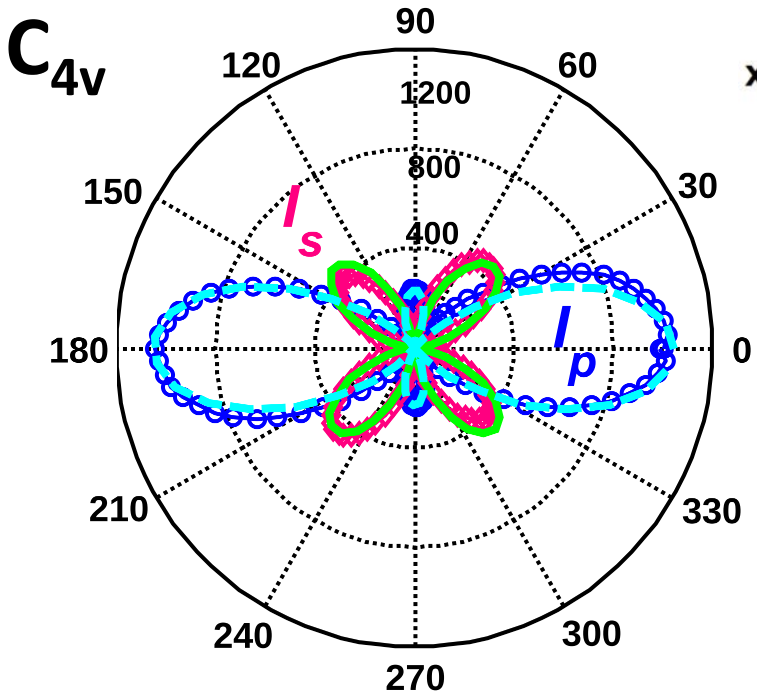


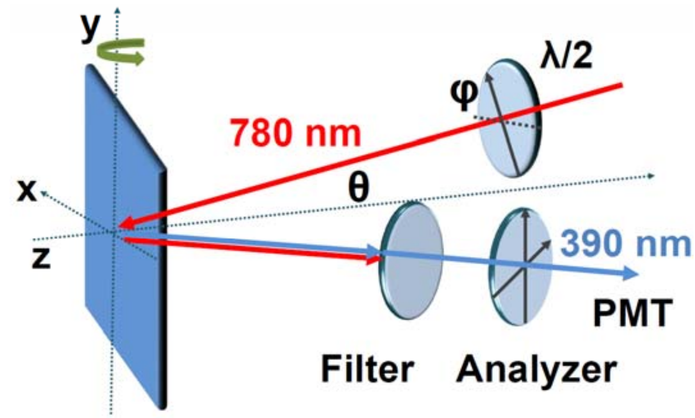
Cartoon courtesy of Müller, *et al* , NMat 8, 56 (2009) ; data of Sheu, unpublished

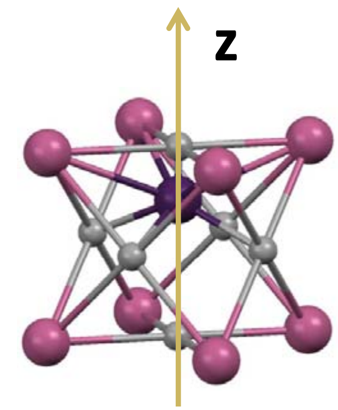
Idea of ultrafast ME coupling at an interface



SHG and C_{4v} symmetry



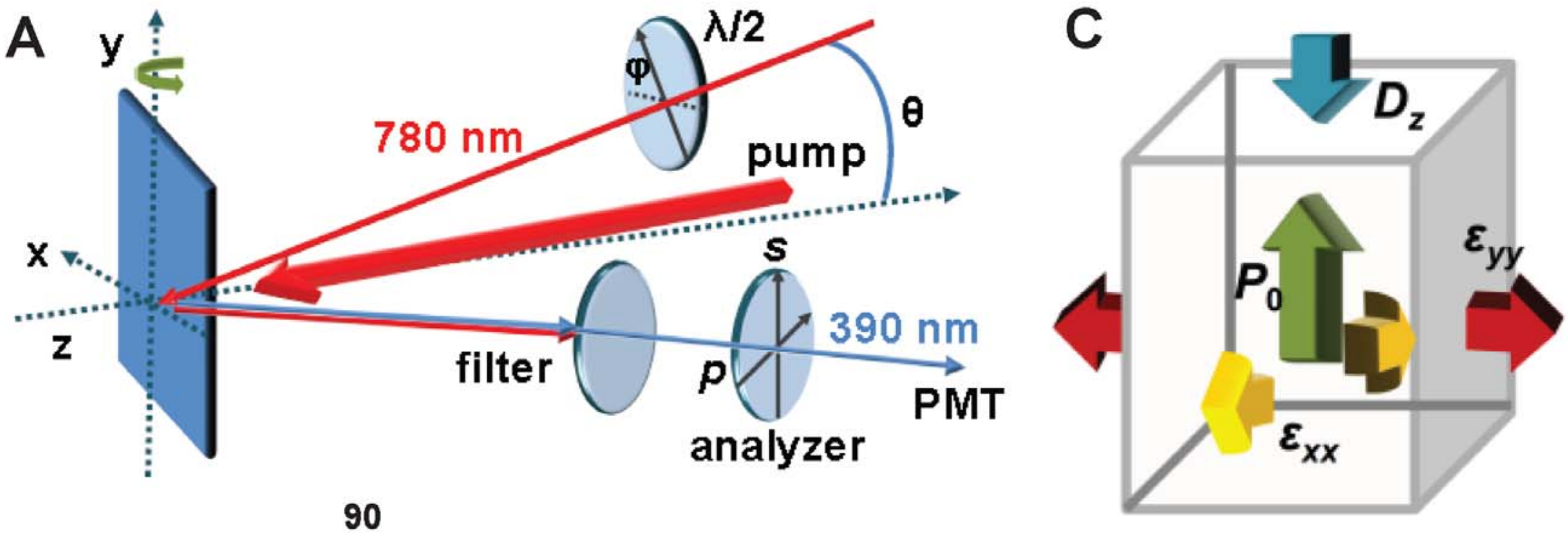




$$E_x = \cos\phi\cos\theta$$
$$E_y = \sin\phi$$
$$E_z = -\cos\phi\sin\theta$$

$$\begin{pmatrix} P_x \\ P_y \\ P_z \end{pmatrix} = \begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{15} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_{1x}E_{2x} \\ E_{1y}E_{2y} \\ E_{1z}E_{2z} \\ E_{1y}E_{2z} \\ E_{1x}E_{2z} \\ E_{1x}E_{2y} \end{pmatrix}$$

DC, linear optics, and SHG



DC: piezoelectric response

$$D_i = e_{ijk}\varepsilon_{jk} = \chi_{ii}^{0} E_i$$

ω: electro-optical response

$$n_i = (1 + 4\pi\chi_{ii}^{\omega})^{1/2}$$

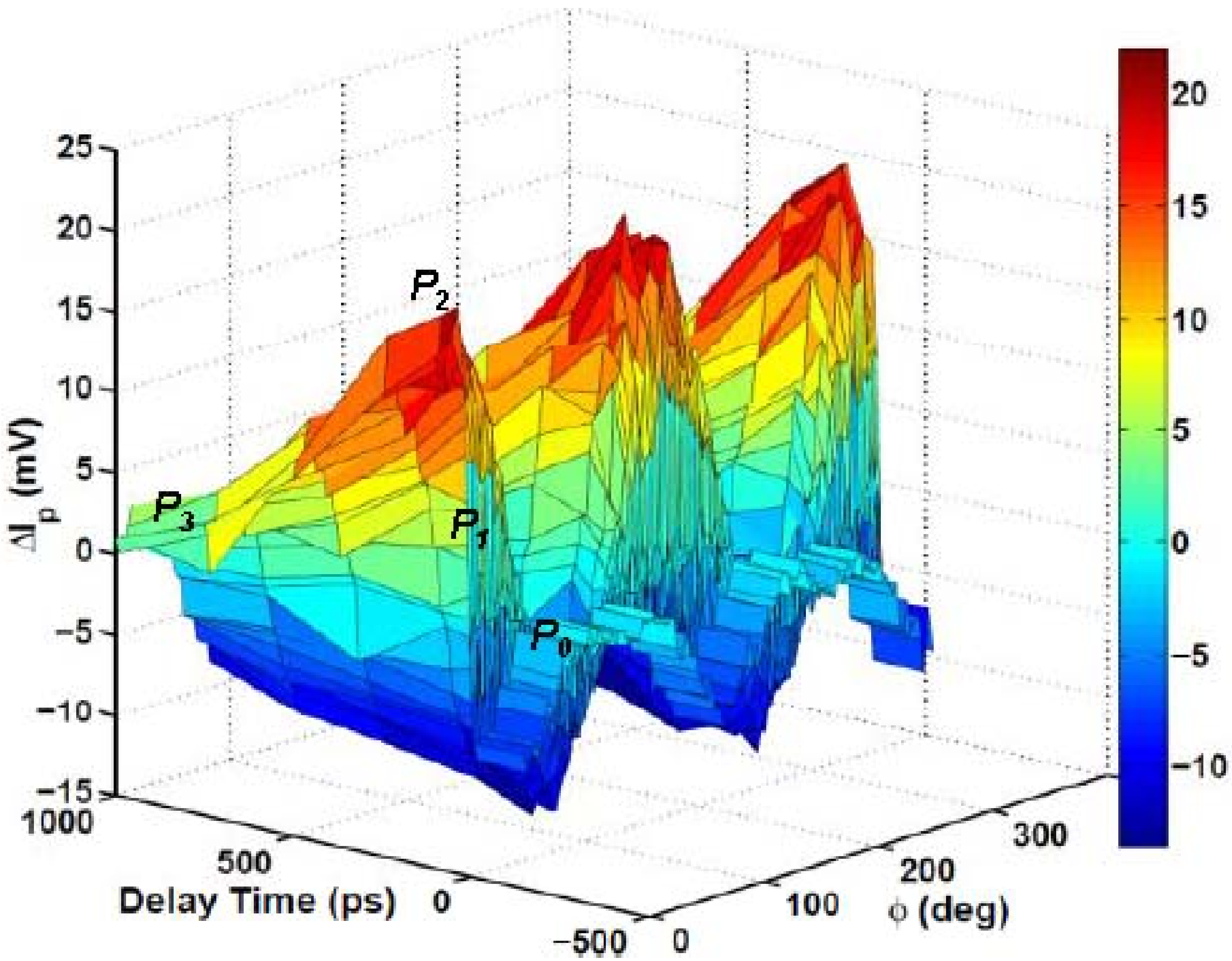
2ω: second harmonic response

$$P_i^{2\omega} = \chi_{ii}^{2\omega} E_i^{2\omega}$$

$$= \chi_{ii}^{2\omega}\delta_{ijk} P_j P_k$$

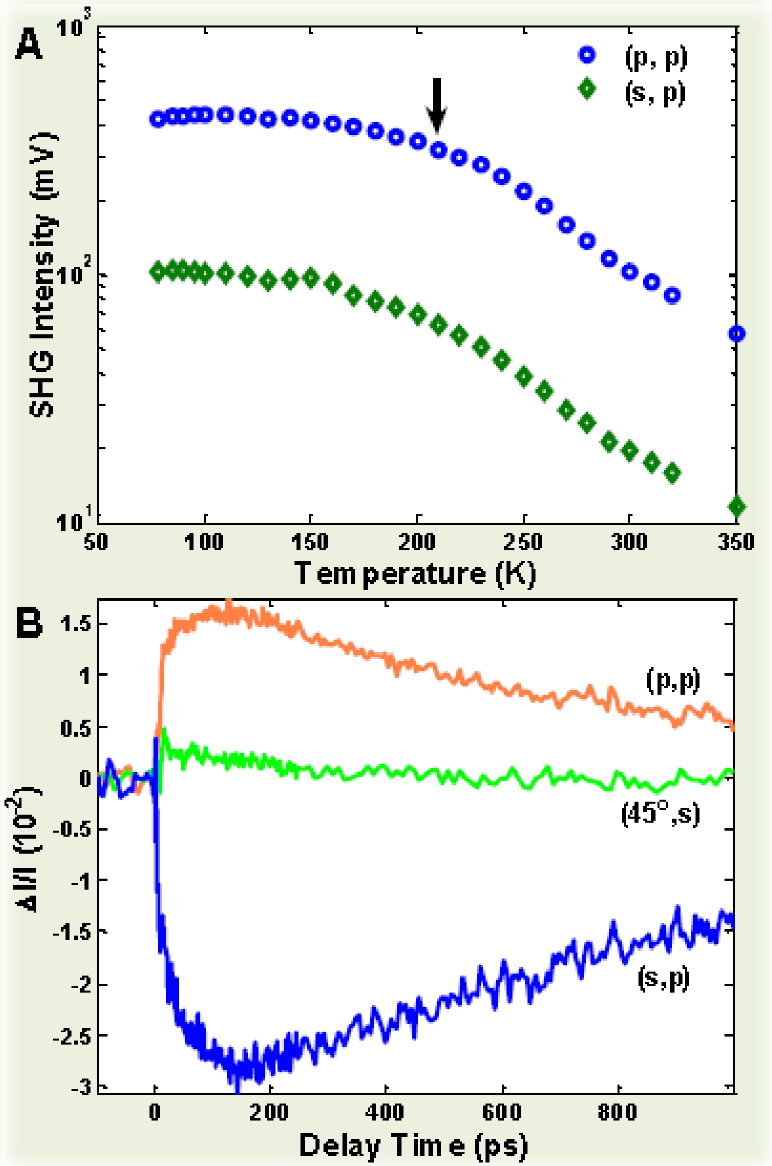
$$= \chi_{ii}^{2\omega}\delta_{ijk}\chi_{jj}^{\omega} E_j \chi_{kk}^{\omega} E_k$$

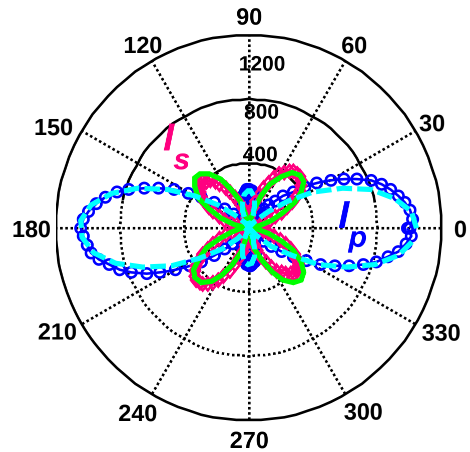
TRSHG upon photoexcitation of LCMO



Sheu, *et al.*, submitted (2014)

Evidence of nonthermal origin





$$I_p = (a\cos^2\phi + b\sin^2\phi)^2,$$
$$I_s = (c\sin(2\phi))^2.$$

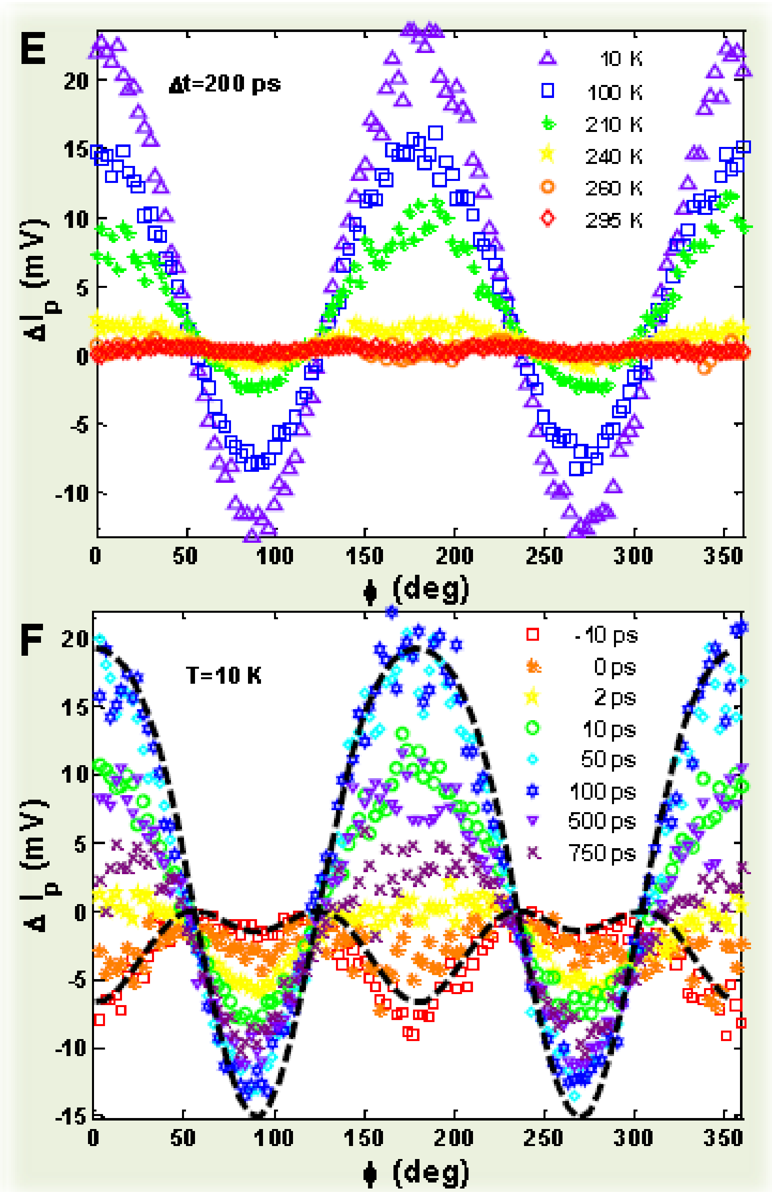
	$(45,p)$	(s,p)	(p,p)
SHG dipole	$P_y(2\omega)$	$P_z(2\omega)$	$P_x(2\omega), P_z(2\omega)$
(a,b,c)	$c{=}f(d_{15})$ >0	$b{=}f(d_{31})$ <0	$a{=}f(d_{15},d_{31},d_{33})$ >0
(d_{15},d_{31},d_{33})	d_{15}=-1.2	d_{31}=-1	$d_{15},d_{31},d_{33}=15$

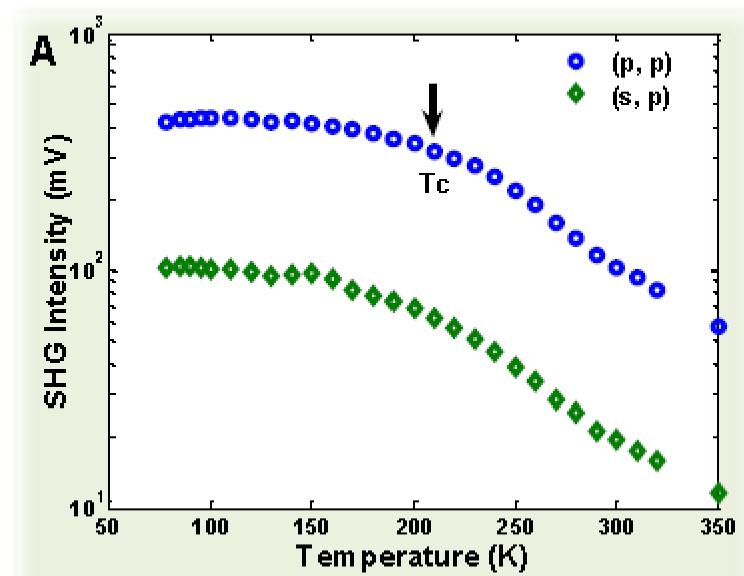
TABLE I: The various polar combinations probe coefficients (a,b,c), $\chi^{(2)}$ component (d_{15},d_{31},d_{33}).Each coefficient (a,b,c) are functions, f, of (d_{15},d_{31},d_{33}).

- **Temperature decreases SHG intensity for all polar combinations**
- **d_{31} and d_{33} changes significantly**

Sheu, *et al.*, submitted (2014)

SHG polar changes





Unipolar and bipolar changes:

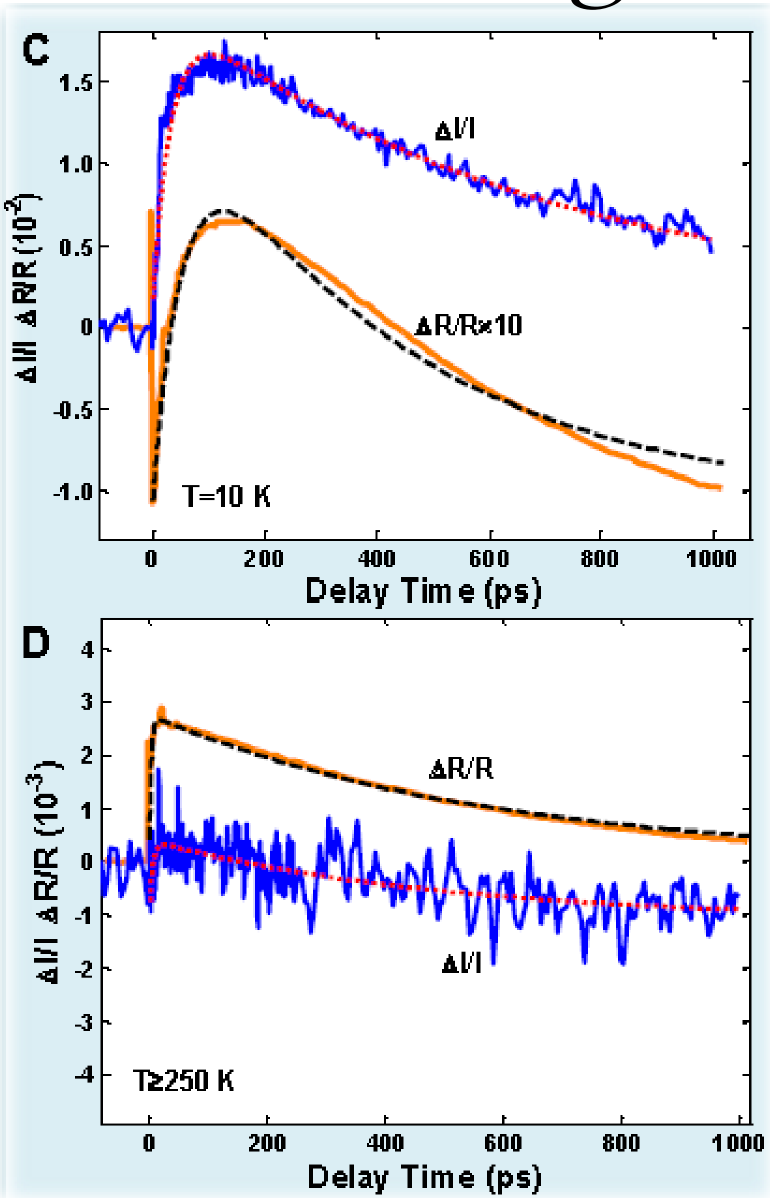
- **Unipolar before t_0: residual heating from 250 kHz laser and slow heat dissipation**

- **Bipolar after t_0 : nonthermal from strain**

- **No phase variation due to symmetry change**

Sheu, *et al.*, submitted (2014)

Evidence of FE change after demagnetization of LCMO



Evidence of nonlinear optical process:

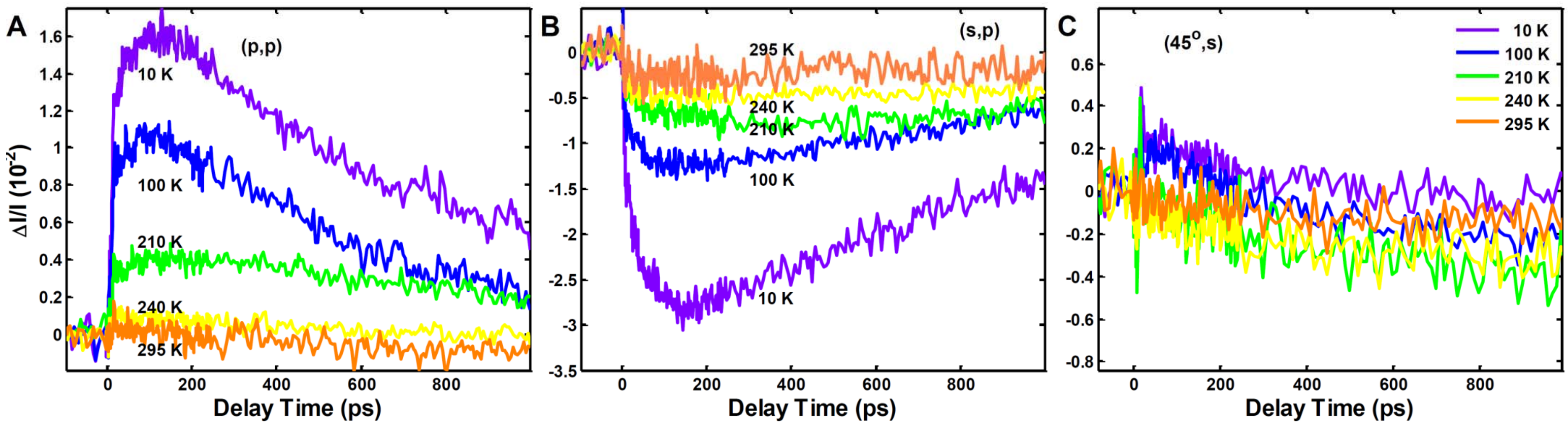
- ΔI/I is one order of magnitude larger
- No polar dependence for ΔR/R
- Not a result from change in optical reflectivity (R)

Evidence of magnetoelastic effect:

- Initial small thermal strain takes 7 ps to propagate across the BSTO film
- Strain relaxation through magnetostriction takes ~50-100 ps
- s-l relaxation channel disappear above Tc
- NO ΔI/I is observed in BSTO "OR" LCMO film

Sheu, *et al.*, submitted (2014)

TRSHG at various temperatures



Interfacial ME coupling:

- **Significant changes are observed in TRSHG below FM T_c**
- **Largest signal at various T below FM follows s-l relaxation time of ~50-100 ps**

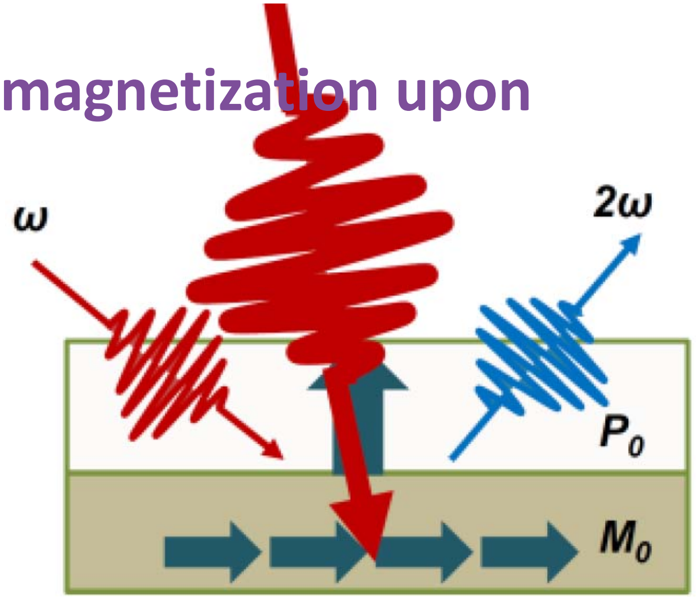
Sheu, *et al.*, submitted (2014)

Conclusion

- Development of using ultrafast optical methods to induce ME coupling by initiating strain relaxation through magnetostriction and probing the piezoelectric response through TRSHG.

- Discovery of timescale governing interfacial ME coupling. Largest piezoelectric response follows s-l relaxation.

- Evidence of FE change in BSTO after demagnetization upon photoexcitation in LCMO.



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