Photo-creating novel light-matter interaction

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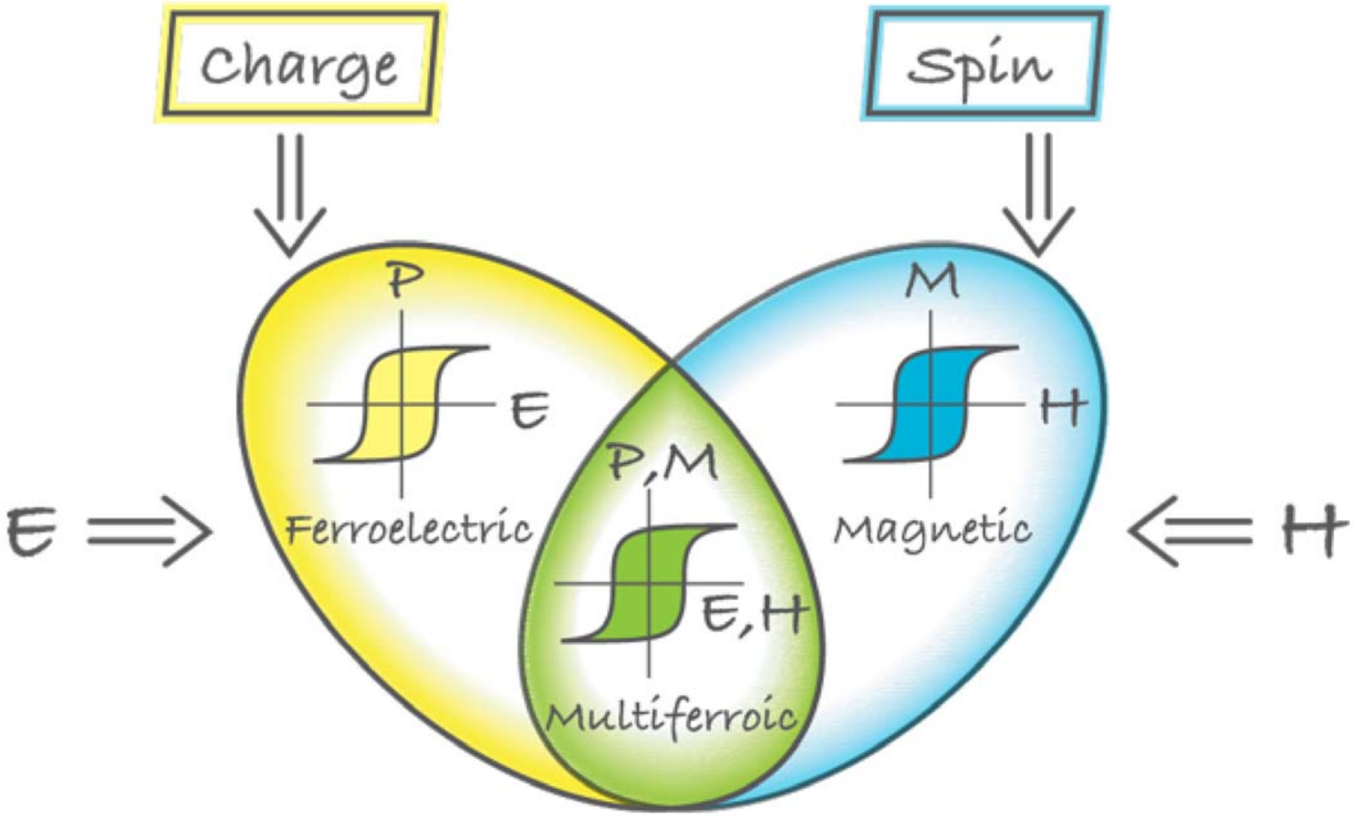
Outline

- Multiferroics and classification of
  1. Proper ferroelectrics
  2. Improper ferroelectrics
- Why multiferroics? How to manipulate ferroelectric dipoles and magnetic spins using ultrafast lasers? What timescale?
- Indirect coupling at interface of composite multiferroics : $BaSrTiO_3/La_{0.7}Sr_{0.3}MnO_3$
- Direct coupling through spin spiral in $RMnO_3$
- Future: coherent control?

Magnetoelectric Mutiferroics

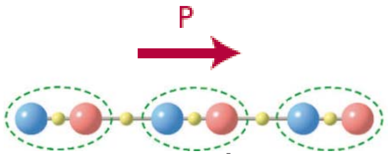


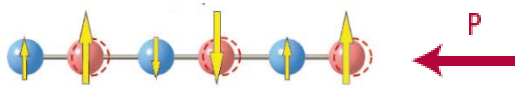
Cartoon courtesy of Khomskii , Physics 2, 20 (2009)

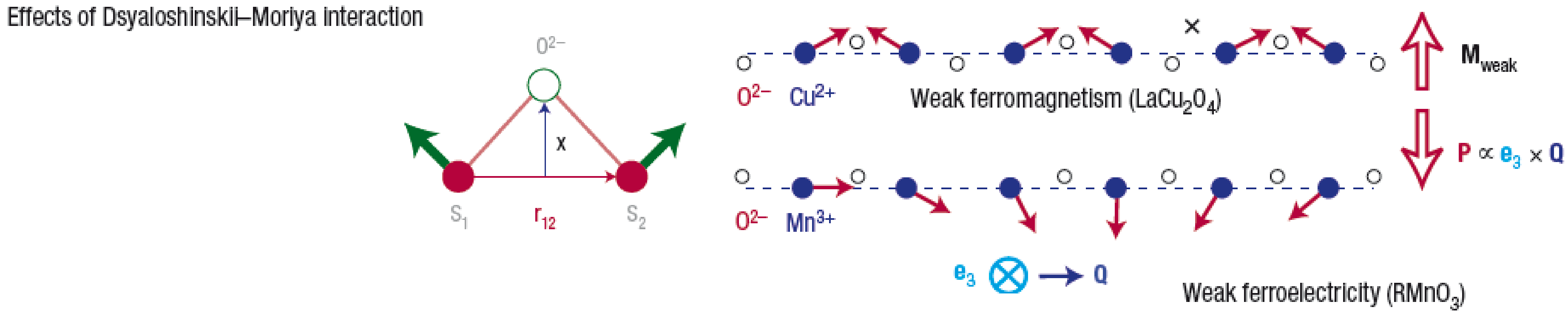
Classification of ferroelectrics

Table 1 Classification of ferroelectrics — Cheong et al., Nature 6, 13 (2007)

	Mechanism of inversion symmetry breaking	Materials
Proper	Covalent bonding between $3d^0$ transition metal (Ti) and oxygen	$BaTiO_3$
	Polarization of $6s^2$ lone pair of Bi or Pb	$BiMnO_3$, $BiFeO_3$, $Pb(Fe_{2/3}W_{1/3})O_3$
Improper	Structural transition 'Geometric ferroelectrics'	K_2SeO_4, Cs_2CdI_4 hexagonal $RMnO_3$
	Charge ordering 'Electronic ferroelectrics'	$LuFe_2O_4$
	Magnetic ordering 'Magnetic ferroelectrics'	Orthorhombic $RMnO_3$, RMn_2O_5, $CoCr_2O_4$

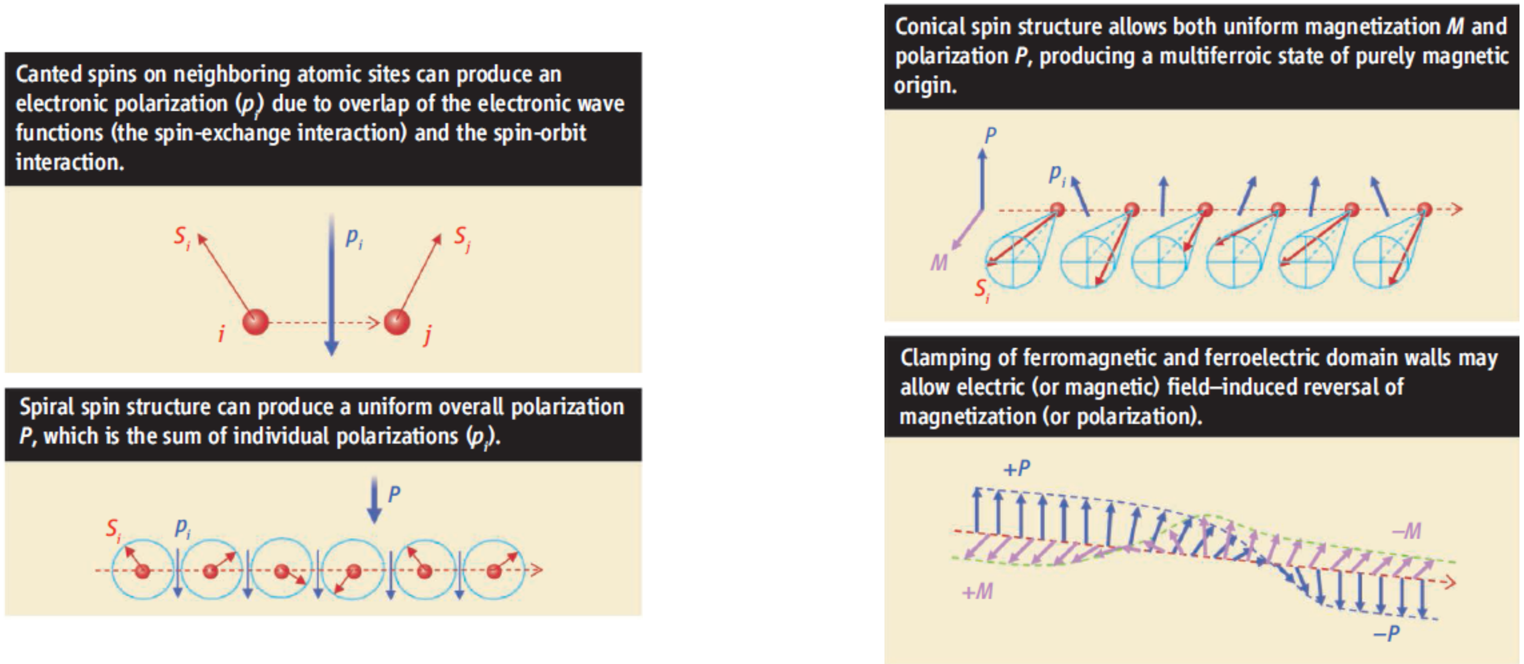






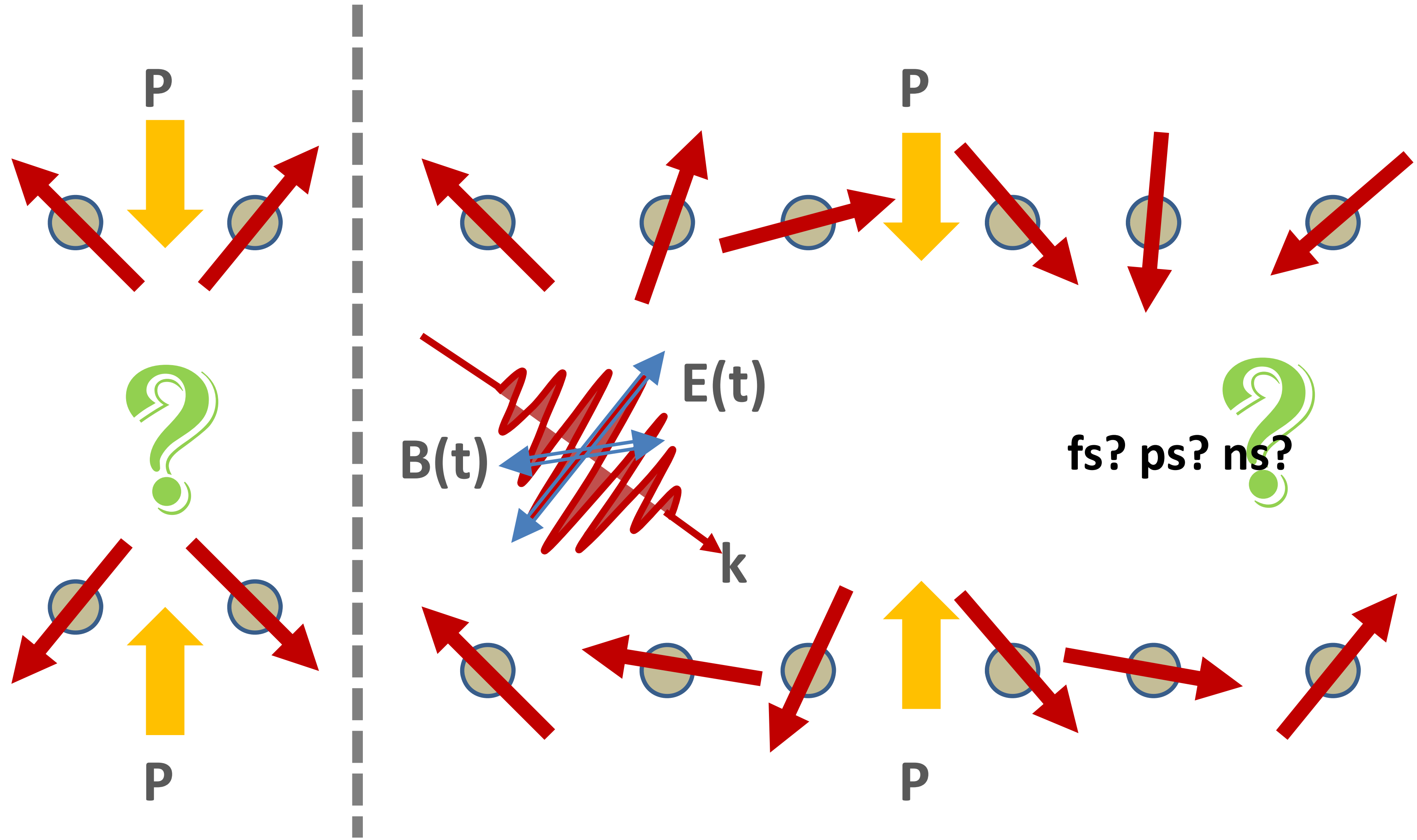
Quantum Electromagnets

Possible spin superstructure with strong ME coupling :

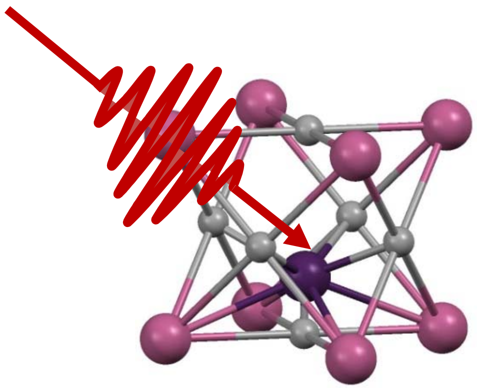


Y. Tokura, Science 312, 1481 (2006)

How quick can one couple E/M?



Ultrafast light sources



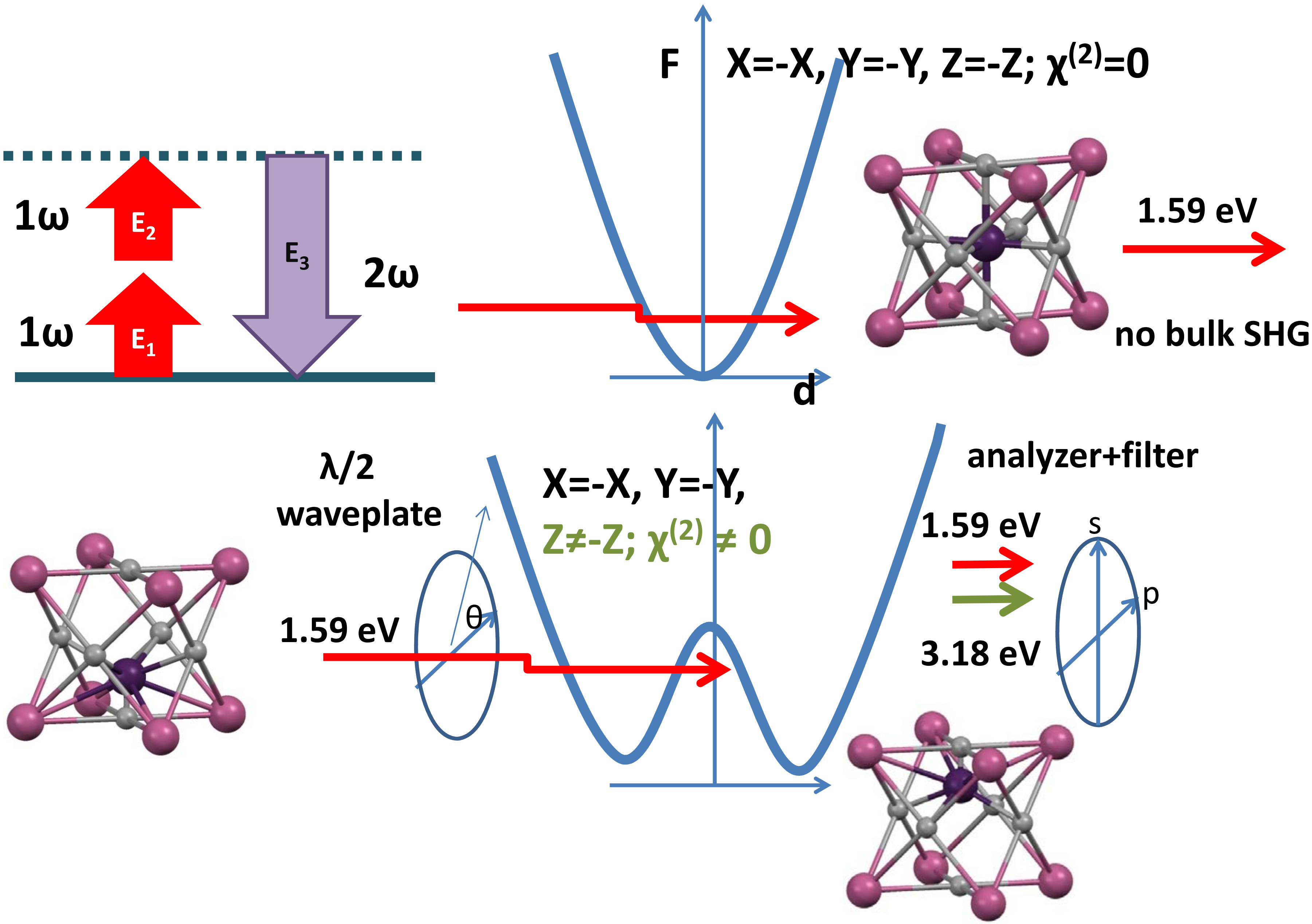
Optical frequency mixing → **Spectroscopy**

Second harmonic generation → **Surface probe, domain imaging**
Crystal symmetry studies

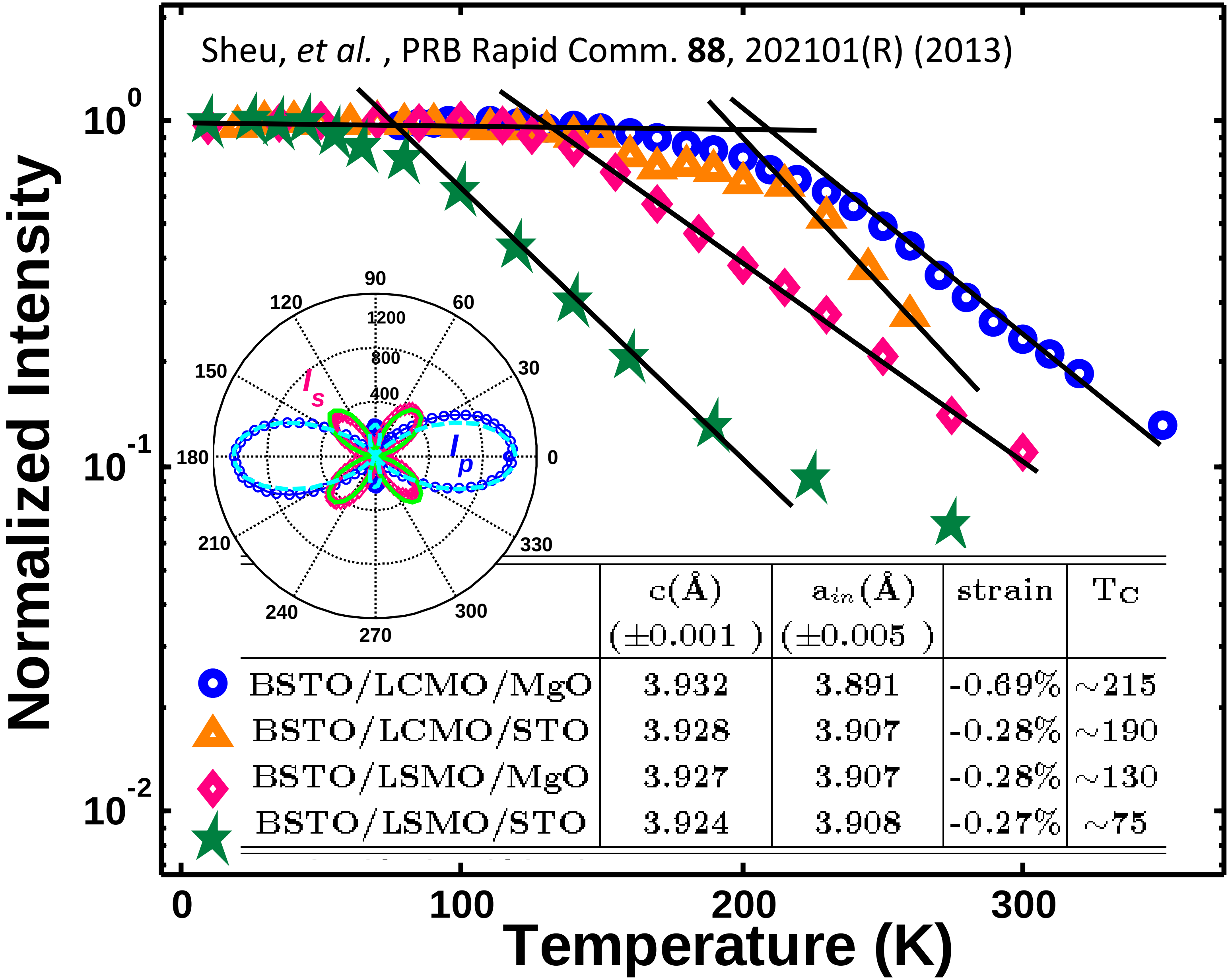
High harmonic generation
X-UV, x rays… → **Element sensitive probes,**
Imaging, diffractions

THz generation → **Quasi-particle excitation and detection, meta-materials**

SHG is a good probe of FE order parameter



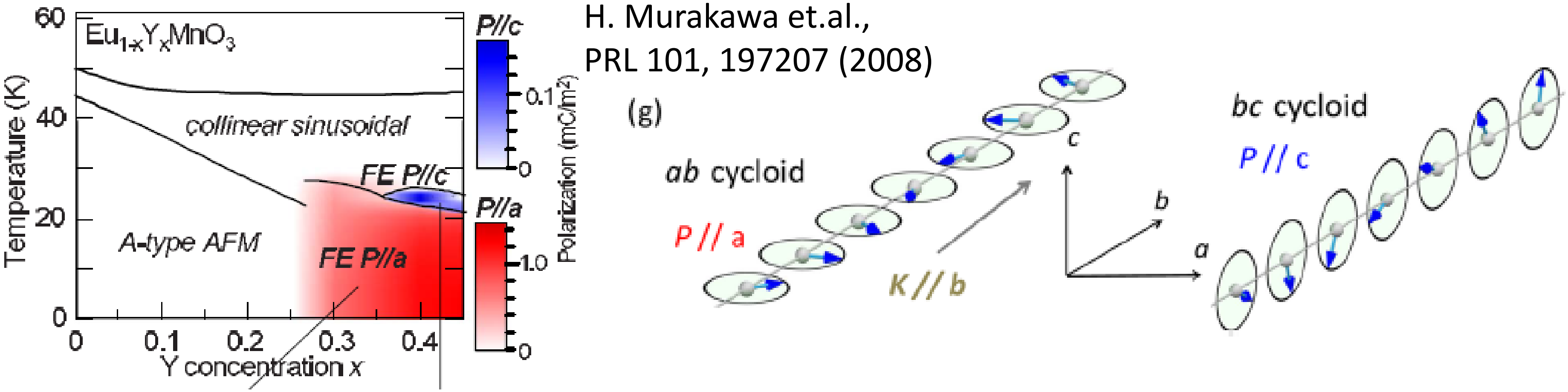
SHG probes FE order: BSTO (FE) and strain

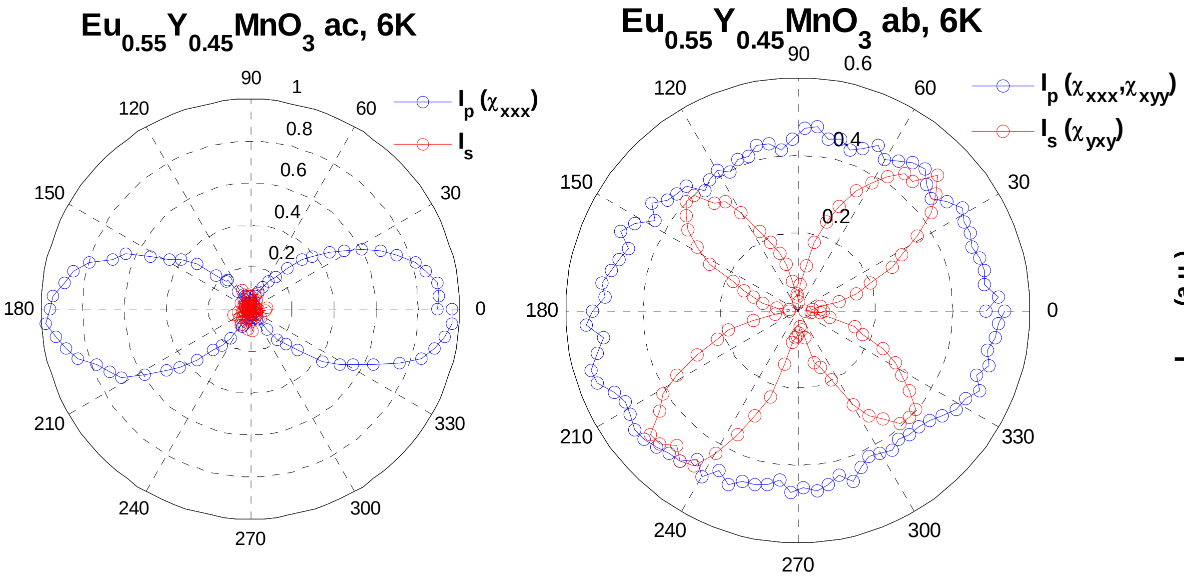


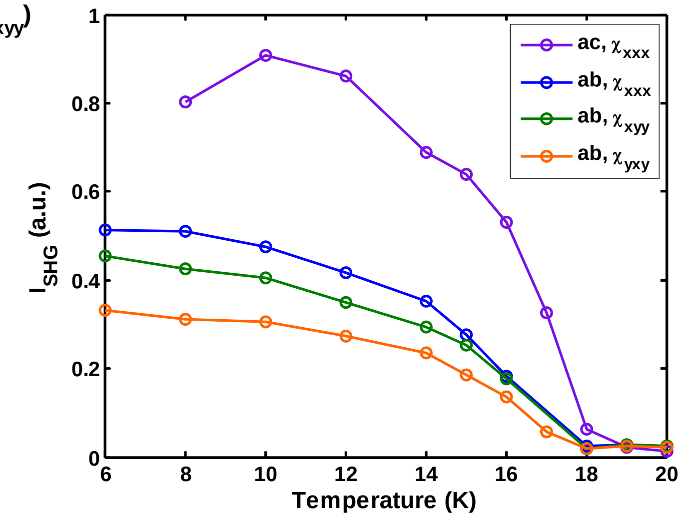
	c(Å) (±0.001)	a_{in}(Å) (±0.005)	strain	T_C
BSTO/LCMO/MgO	3.932	3.891	-0.69%	~215
BSTO/LCMO/STO	3.928	3.907	-0.28%	~190
BSTO/LSMO/MgO	3.927	3.907	-0.28%	~130
BSTO/LSMO/STO	3.924	3.908	-0.27%	~75

SHG probes FE orders: $RMnO_3$

H. Murakawa et.al.,
PRL 101, 197207 (2008)







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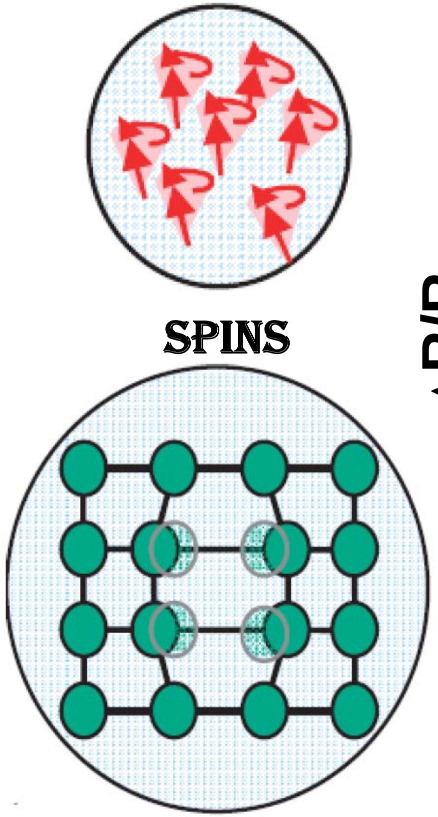
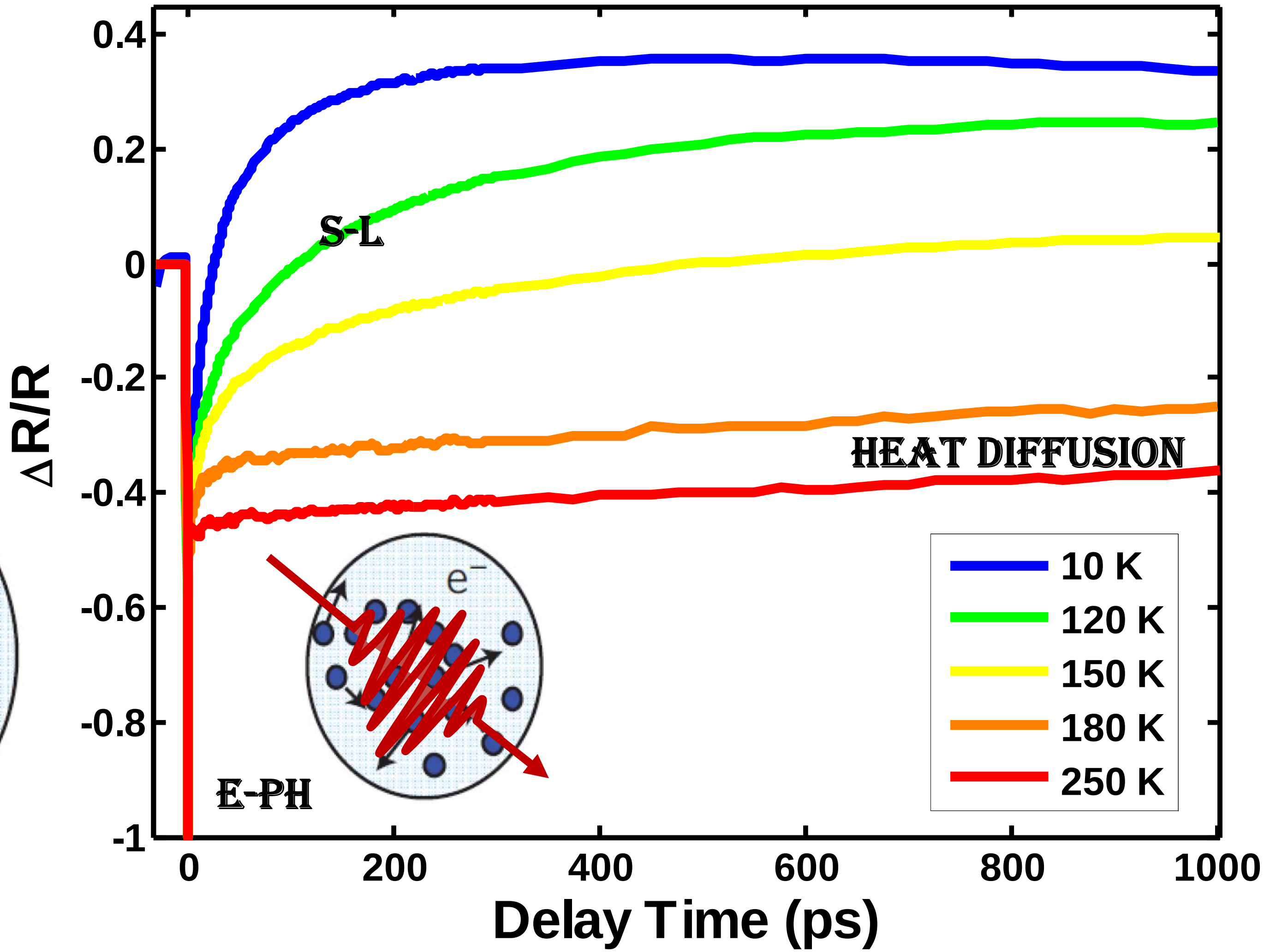
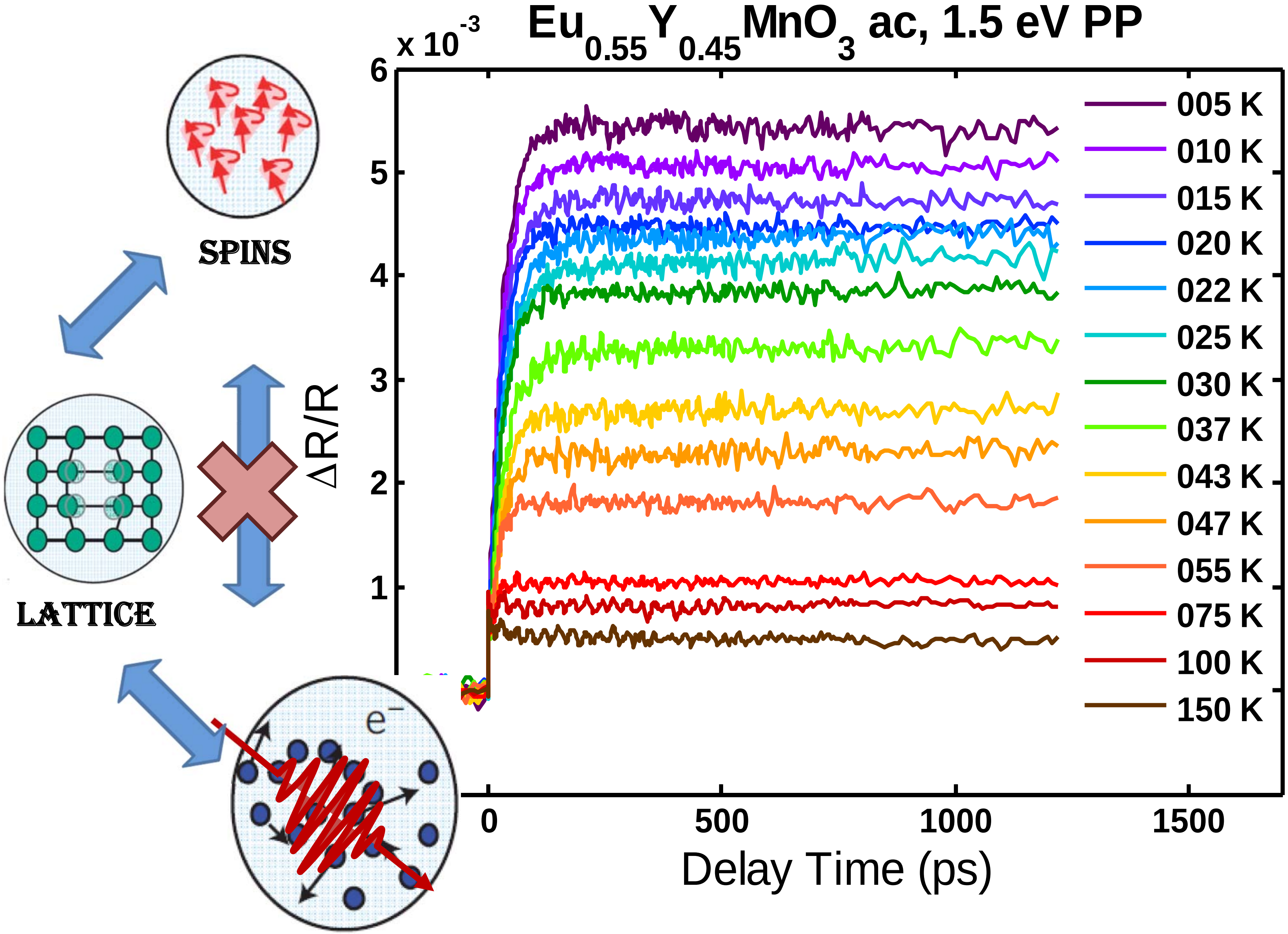
Quasiparticle dynamics of $RMnO_3$





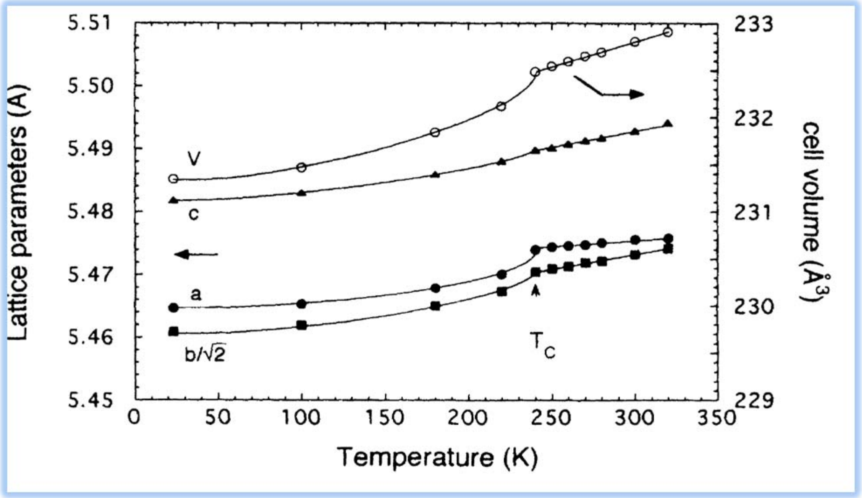
Cartoon courtesy of Müller*, et al*., NMat 8, 56 (2009) ; data of Sheu, unpublished



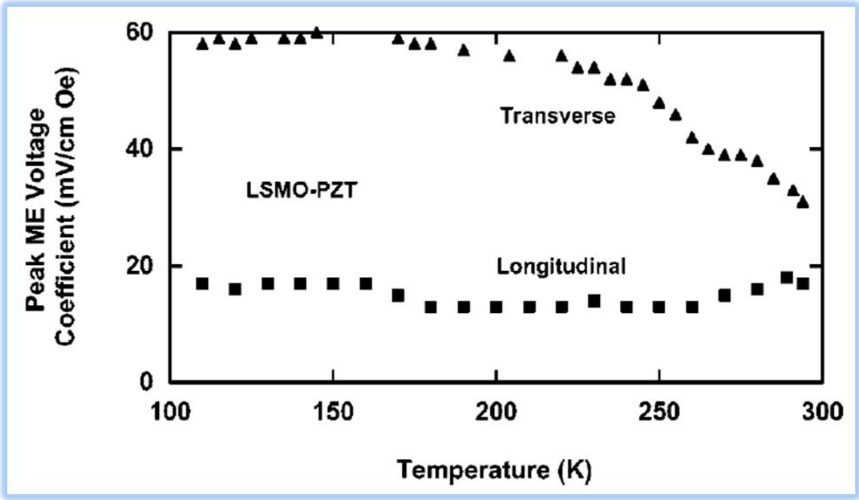
SPINS
LATTICE
e⁻
$Eu_{0.55}Y_{0.45}MnO_3$ ac, 1.5 eV PP
x 10^-3
ΔR/R
Delay Time (ps)
0
500
1000
1500
1
2
3
4
5
6
005 K
010 K
015 K
020 K
022 K
025 K
030 K
037 K
043 K
047 K
055 K
075 K
100 K
150 K

Sheu *et al.*

Magnetostric and piezoelectric effects



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.**

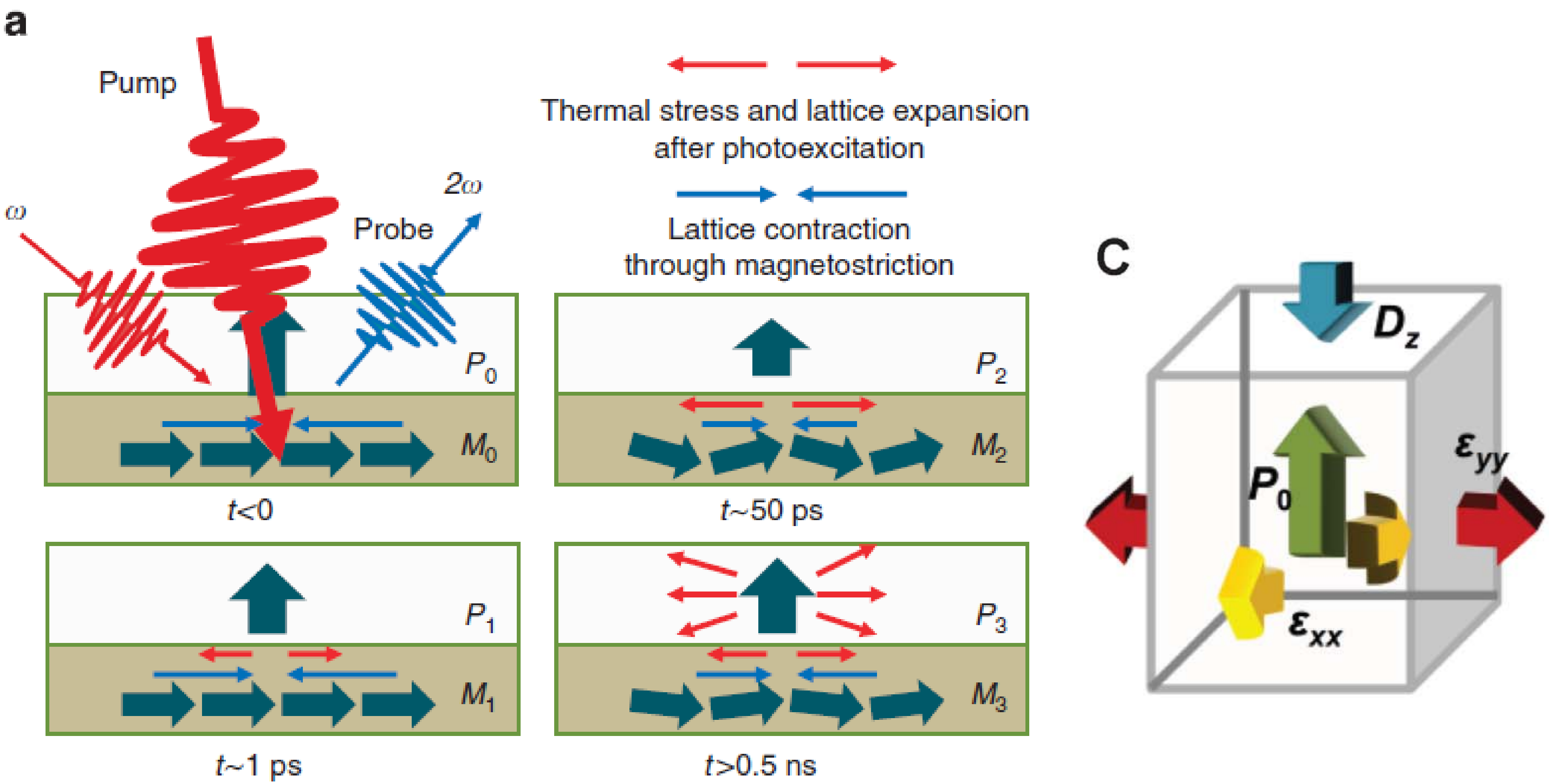
- **Static ME coupling has been achieved in a FM/FE heterostructure of LSMO/PZT via applying an external B field.**

- **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 $T_{c.}$**

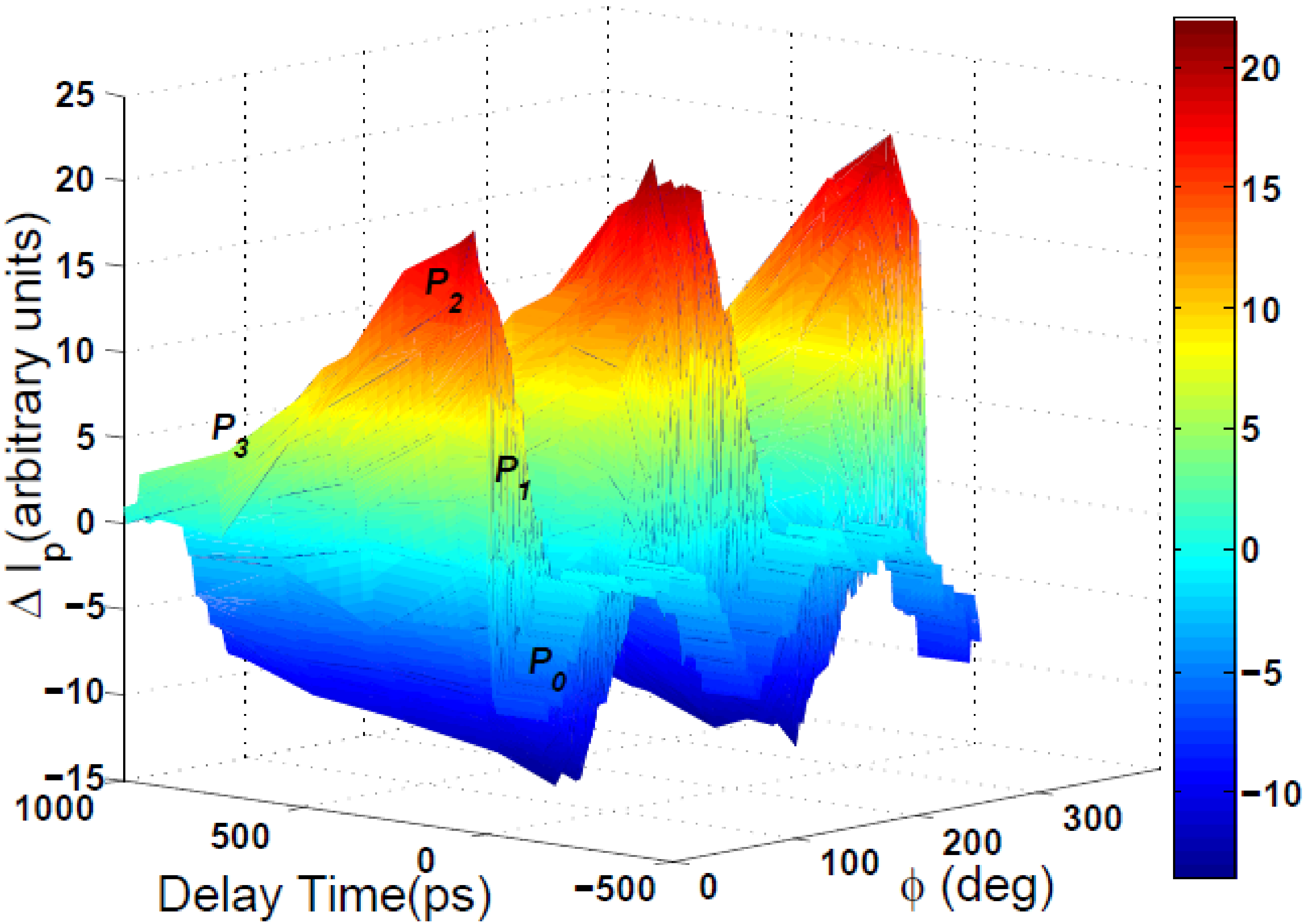
LC(S)MOs are good candidates of magnetostrictive materials.

Ultrafast ME coupling at an interface



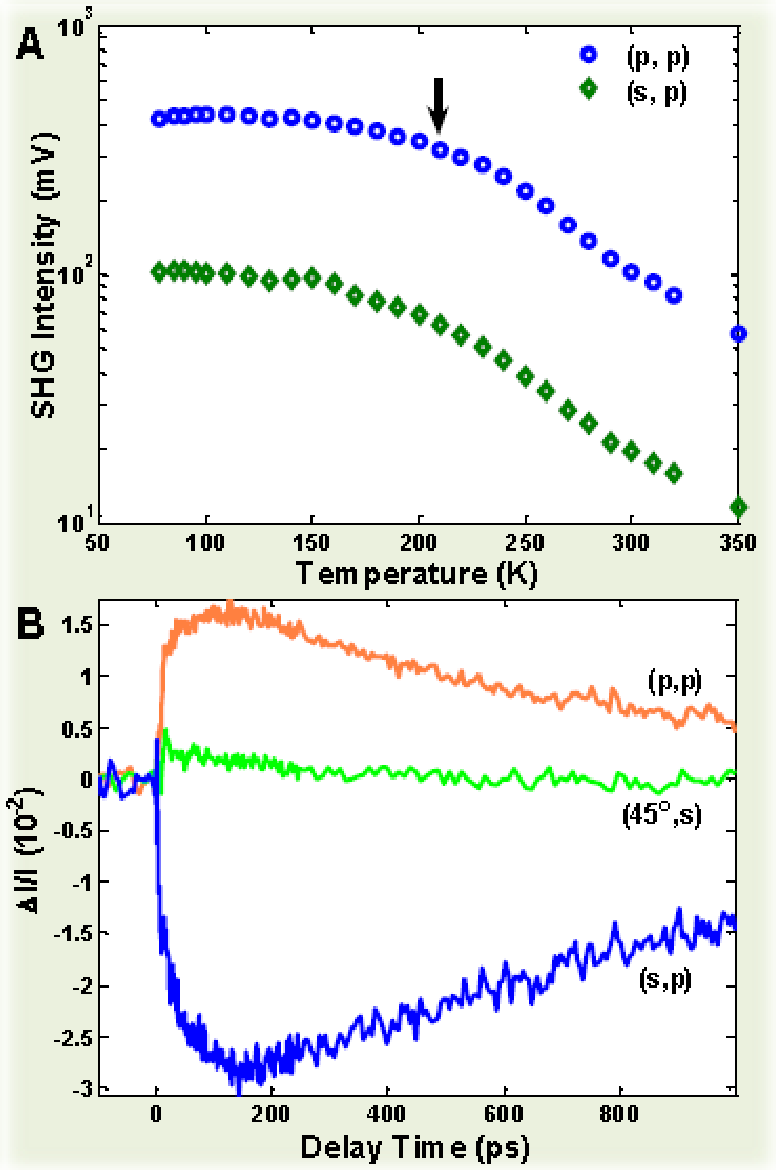
Sheu, *et al.*, Nat. Commun. 5:5832 (2014)

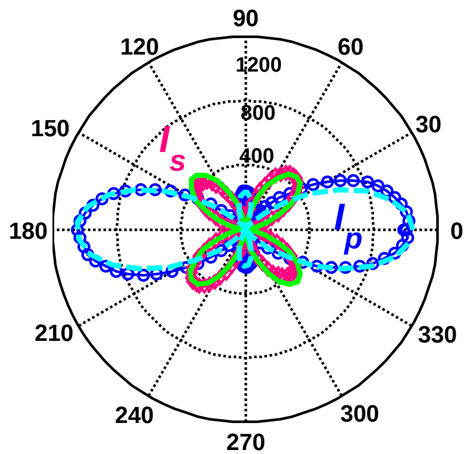
TRSHG upon photoexcitation of LCMO



Sheu, *et al.*, Nat. Commun. 5:5832 (2014)

Evidence of nonthermal origin





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

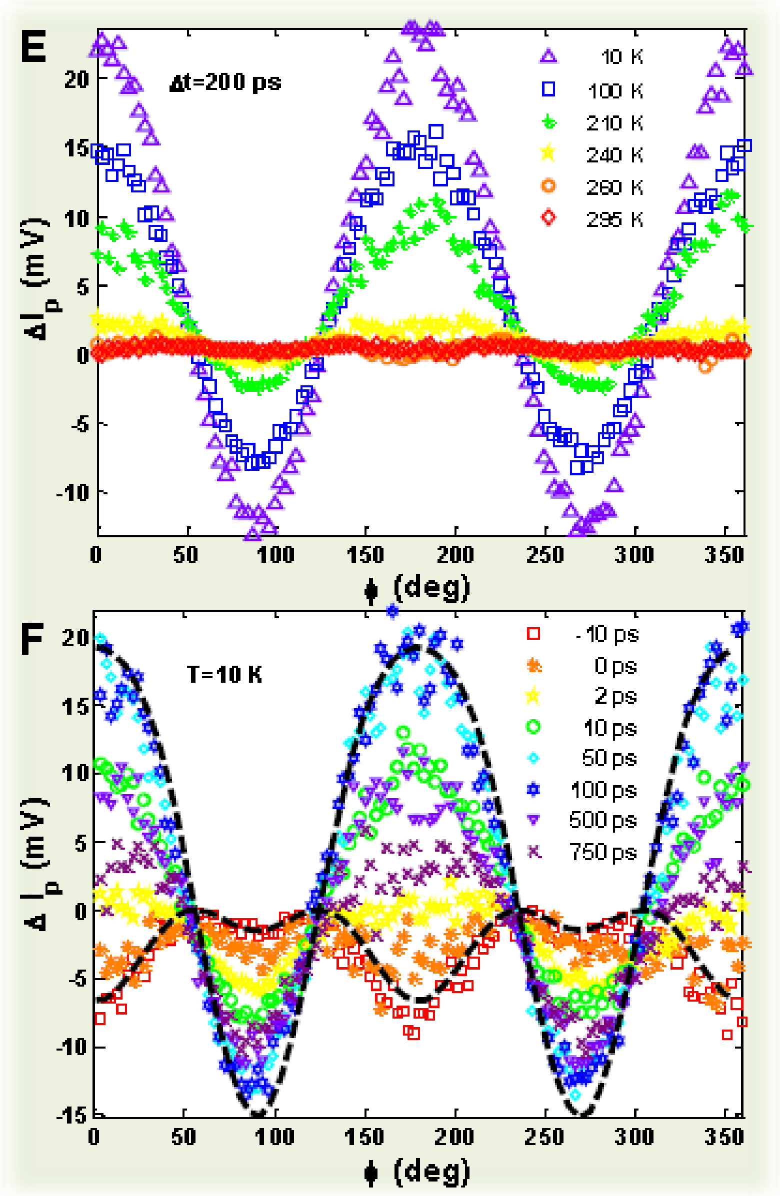
	(45,*p*)	(*s*, *p*)	(*p*, *p*)
SHG dipole	$P_y(2\omega)$	$P_z(2\omega)$	$P_x(2\omega), P_z(2\omega)$
(a, b, c)	$c{=}f(d_{15})$	$b{=}f(d_{31})$	$a{=}f(d_{15}, d_{31}, d_{33})$
	> 0	< 0	> 0
(d_{15},d_{31},d_{33})	d_{15}=-1.2	d_{31}=-1	$d_{15},d_{31},d_{33} = 15$

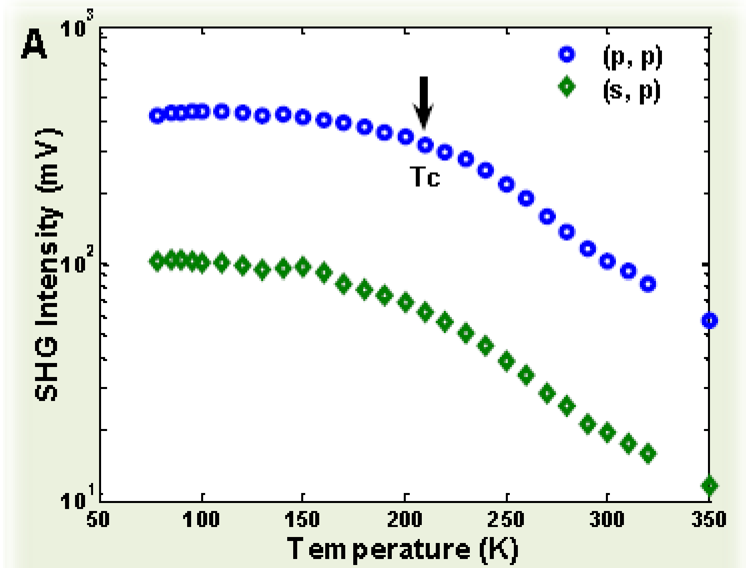
TABLE I: The various polar combinations probe coefficients (a, b, c), $\chi^{(2)}$ component (d_{15},d_{31},d_{33}).Each coefficient (a, b, c) are functions, f, of (d_{15}, d_{31}, d_{33}).

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

Sheu, *et al.*, Nat. Commun. 5:5832 (2014)

SHG polar changes



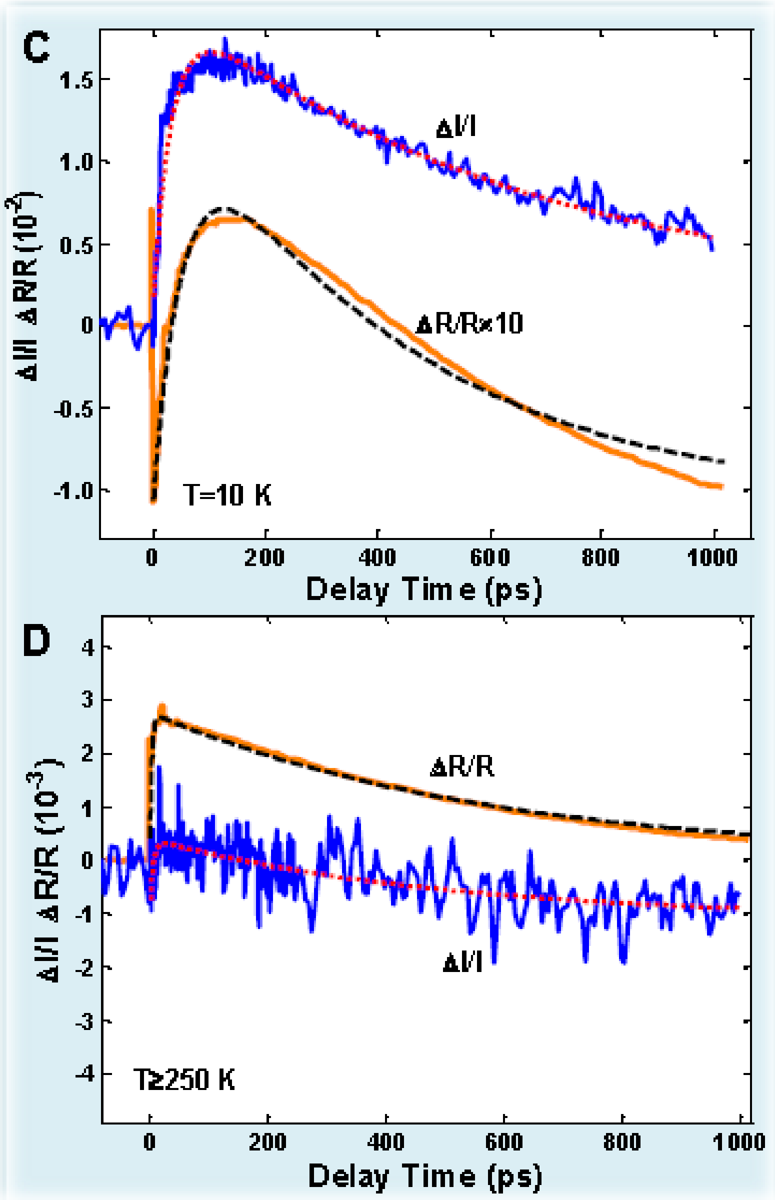


Unipolar and bipolar changes:

- **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.*, Nat. Commun. 5:5832 (2014)

Evidence of FE change after demagnetization of LCMO



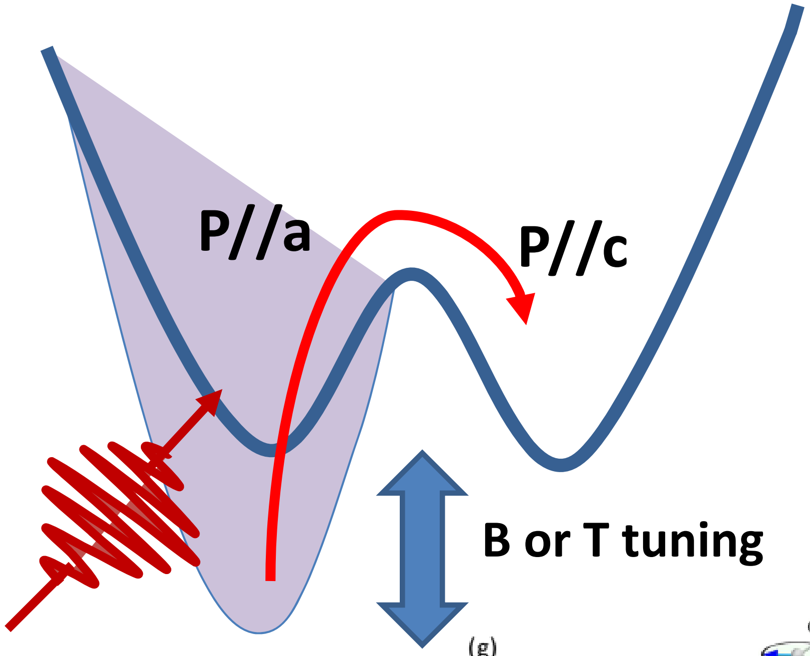
Evidence of nonlinear optical process:

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

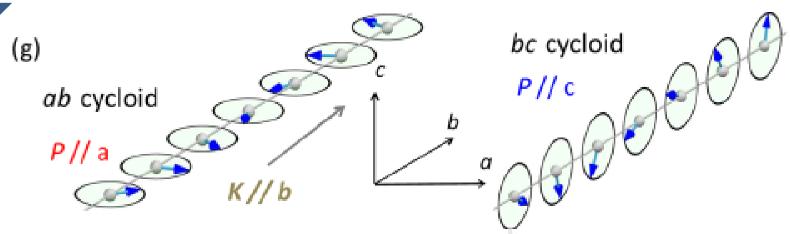
Evidence of magnetoelastic effect:

- **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.*, Nat. Commun. 5:5832 (2014)



H. Murakawa et.al.,
PRL 101, 197207 (2008)



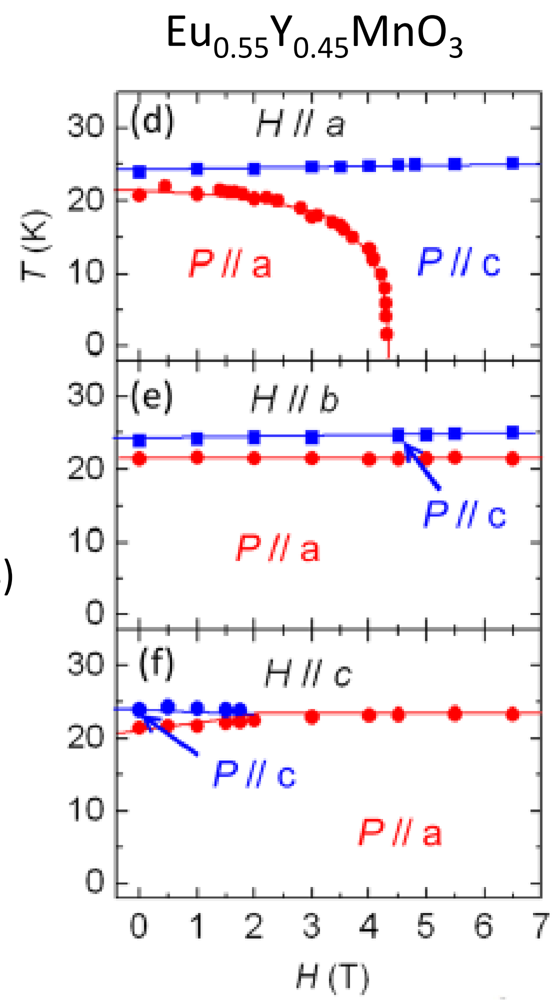
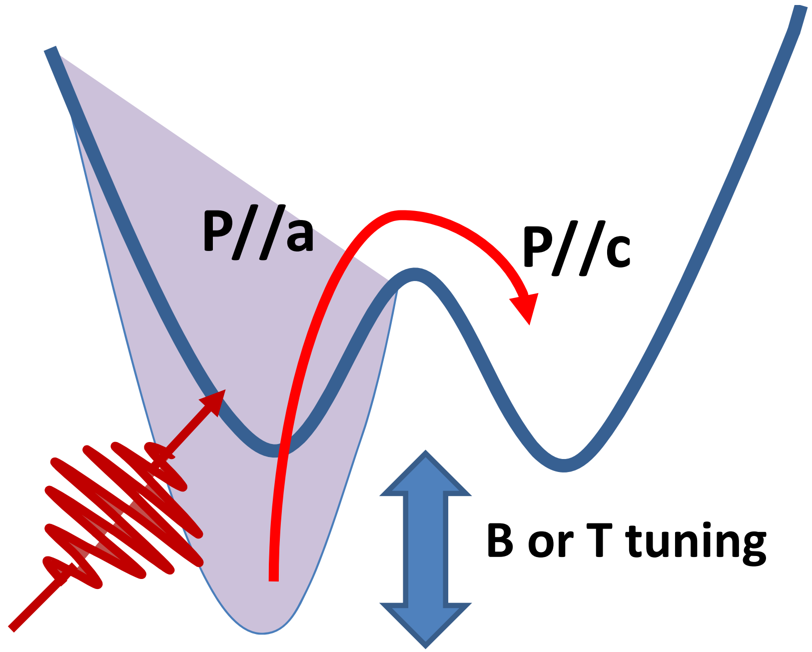
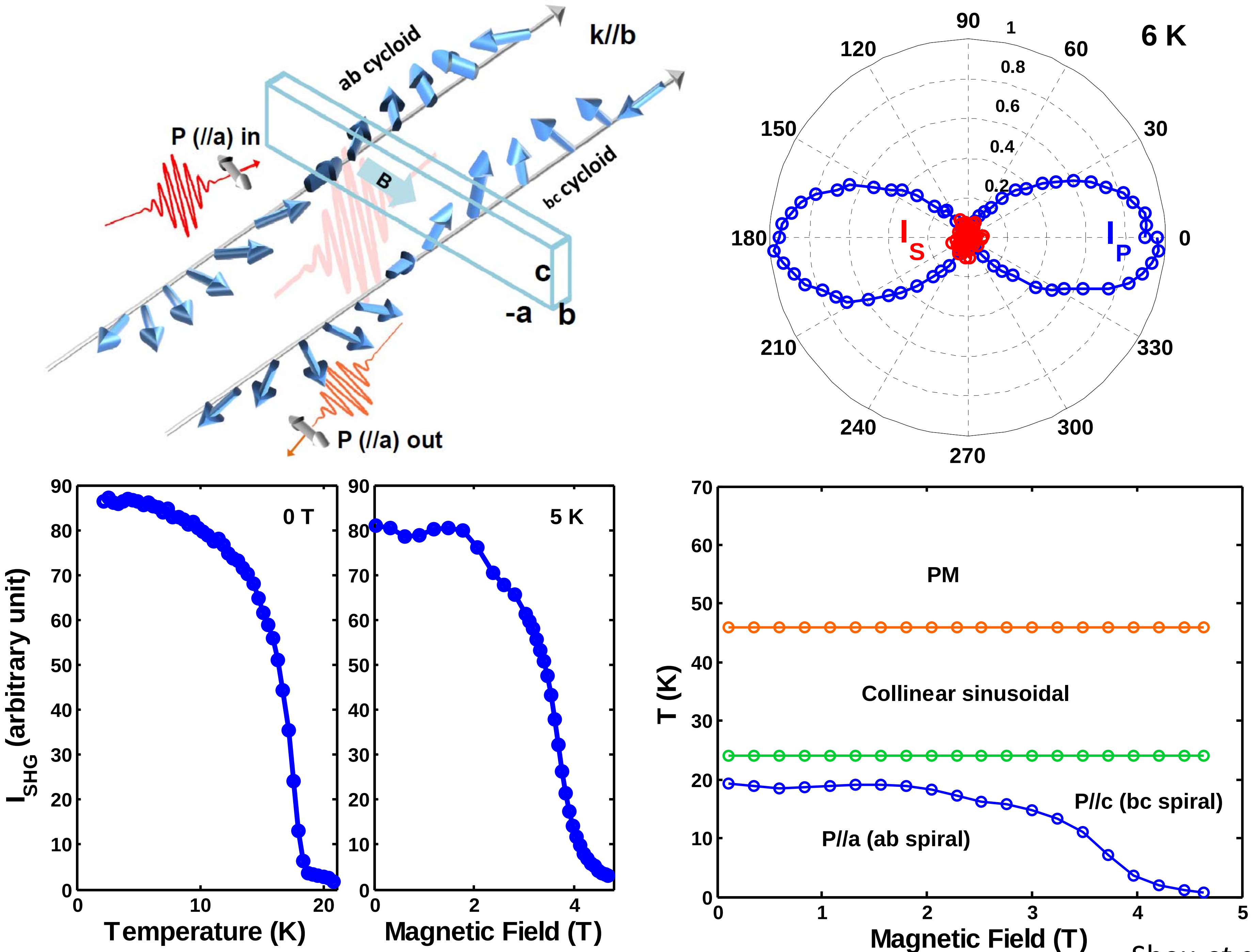


Photo-creating meta-stable states in a spiral-spin multiferroic manganite

Questions to answer:



- AC field of light and what mechanism?
- Critical slowing down?
- Is inertial-driven spin switch between two cycloid states possible by creating collective rotary Nambu-Goldstone mode
- Does the photon directly reconstruct the Mn-spin orientation due to double exchange interaction?
- Can we directly induce a phase transition?


k//b
ab cycloid
P (//a) in
B
bc cycloid
c
-a
b
P (//a) out
6 K
I_S
I_P
0.2
0.4
0.6
0.8
1
0
30
60
90
120
150
180
210
240
270
300
330
I_{SHG} (arbitrary unit)
Temperature (K)
0 T
Magnetic Field (T)
5 K
T (K)
PM
Collinear sinusoidal
P//c (bc spiral)
P//a (ab spiral)
Magnetic Field (T)

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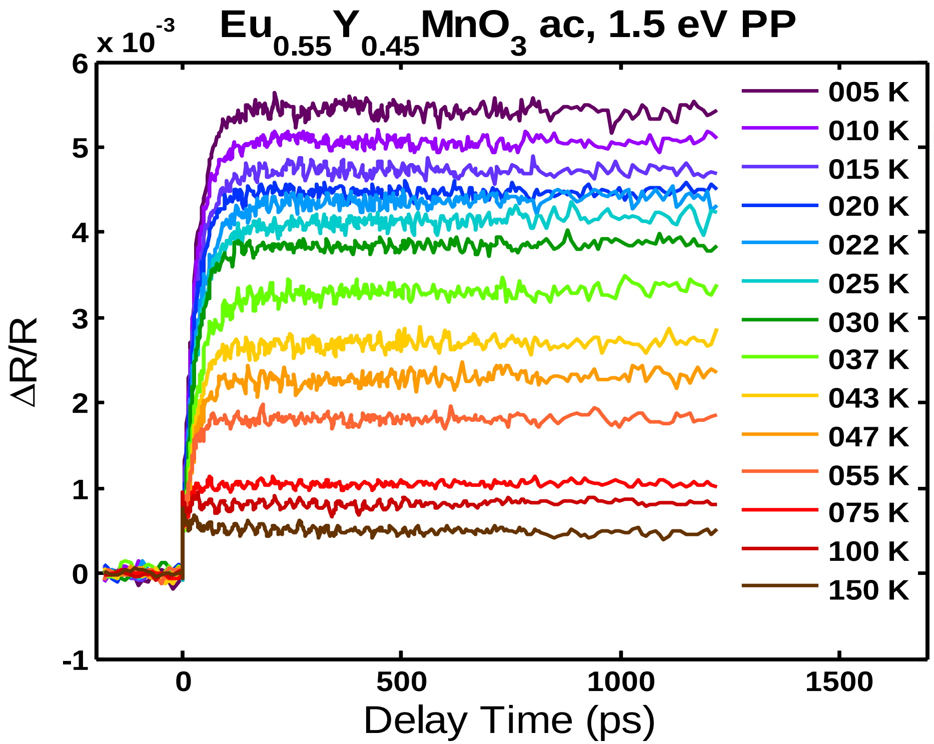
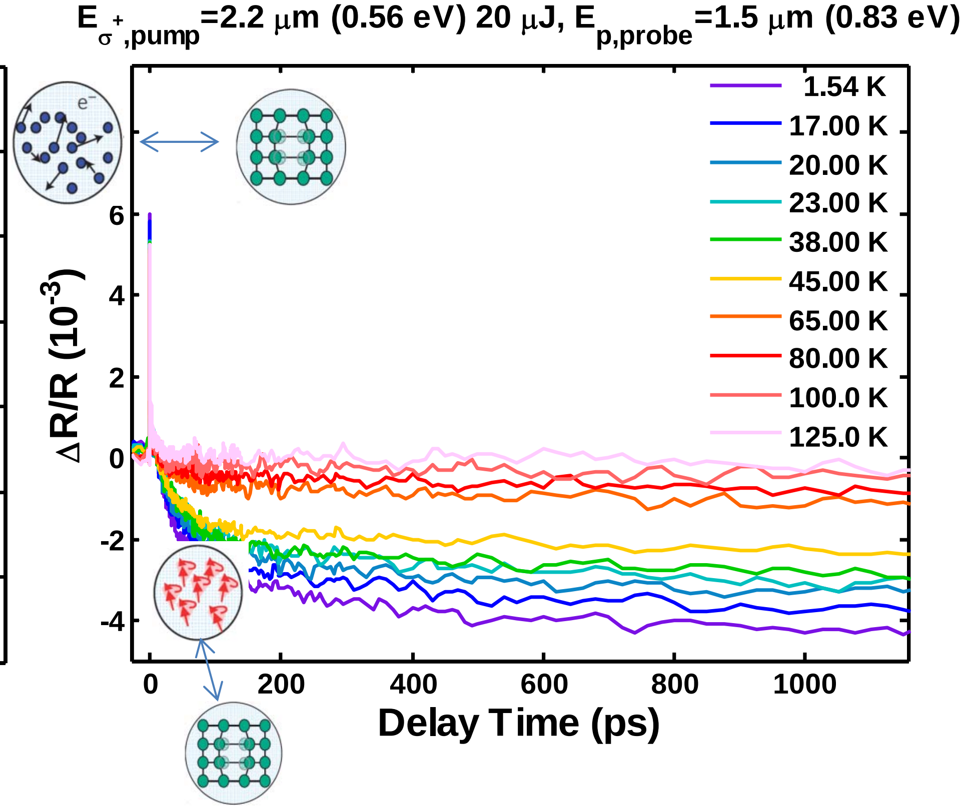
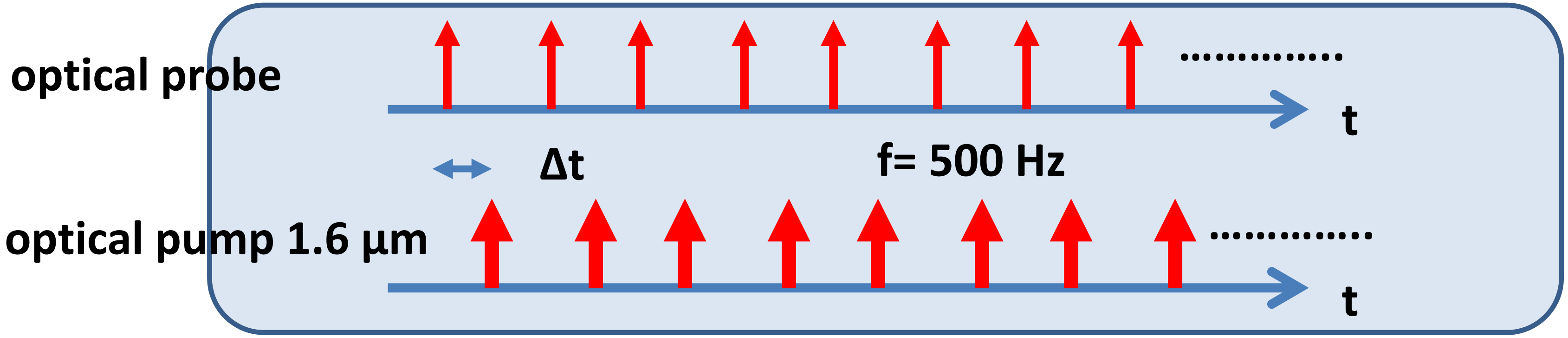
Transient Reflectivity(T,t)

- **Degenerate 1.55v eV pump probe**



- **2-color (nondegenerate) pump probe**





300 ms

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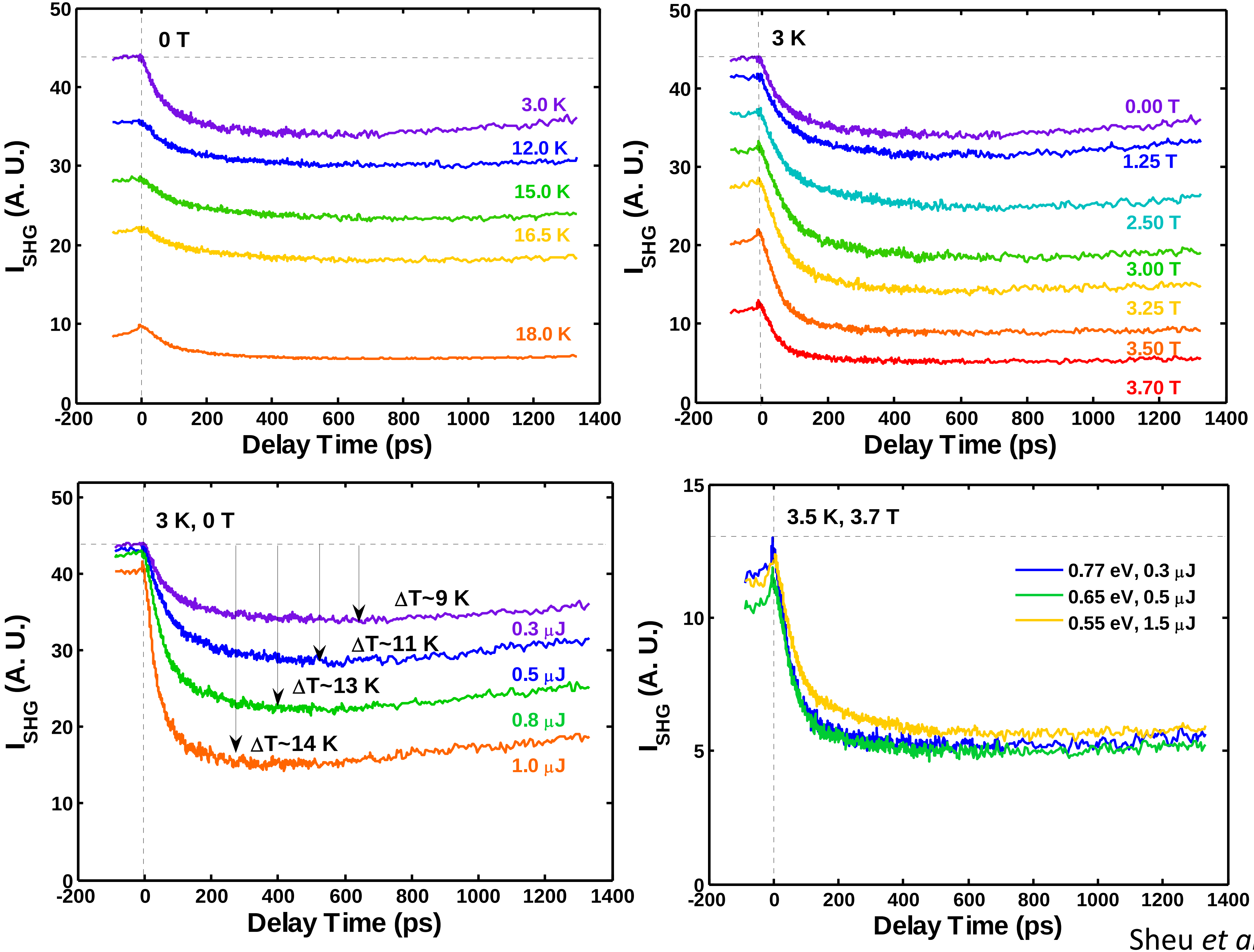

0 T
3.0 K
12.0 K
15.0 K
16.5 K
18.0 K
I_SHG (A. U.)
Delay Time (ps)
3 K
0.00 T
1.25 T
2.50 T
3.00 T
3.25 T
3.50 T
3.70 T
3 K, 0 T
ΔT~9 K
ΔT~11 K
ΔT~13 K
ΔT~14 K
0.3 μJ
0.5 μJ
0.8 μJ
1.0 μJ
3.5 K, 3.7 T
0.77 eV, 0.3 μJ
0.65 eV, 0.5 μJ
0.55 eV, 1.5 μJ

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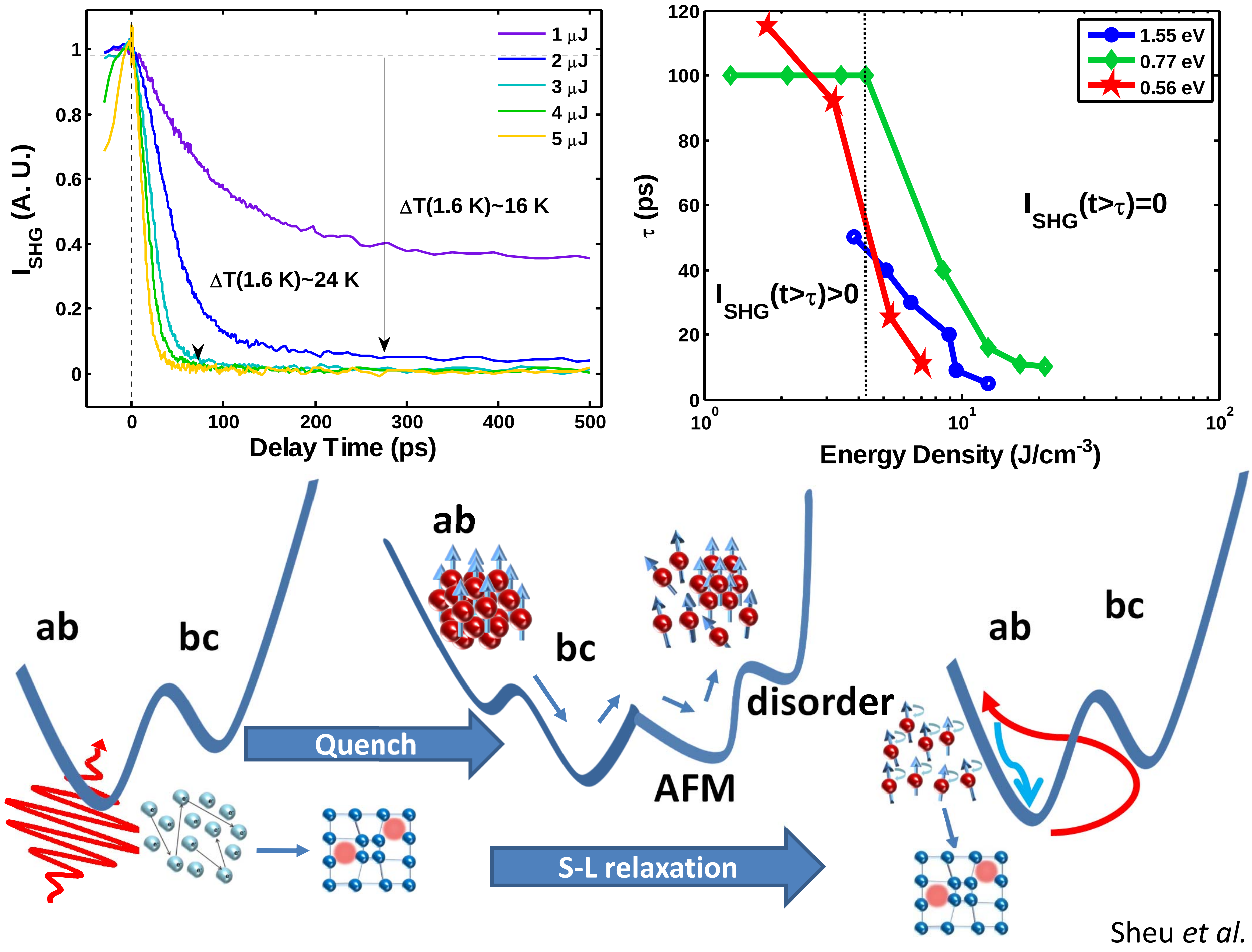

1 μJ
2 μJ
3 μJ
4 μJ
5 μJ
ΔT(1.6 K)~16 K
ΔT(1.6 K)~24 K
I_{SHG} (A. U.)
Delay Time (ps)
1.55 eV
0.77 eV
0.56 eV
$I_{SHG}(t>\tau)=0$
$I_{SHG}(t>\tau)>0$
τ (ps)
Energy Density (J/cm^{-3})
ab
bc
Quench
ab
bc
AFM
disorder
S-L relaxation
ab
bc
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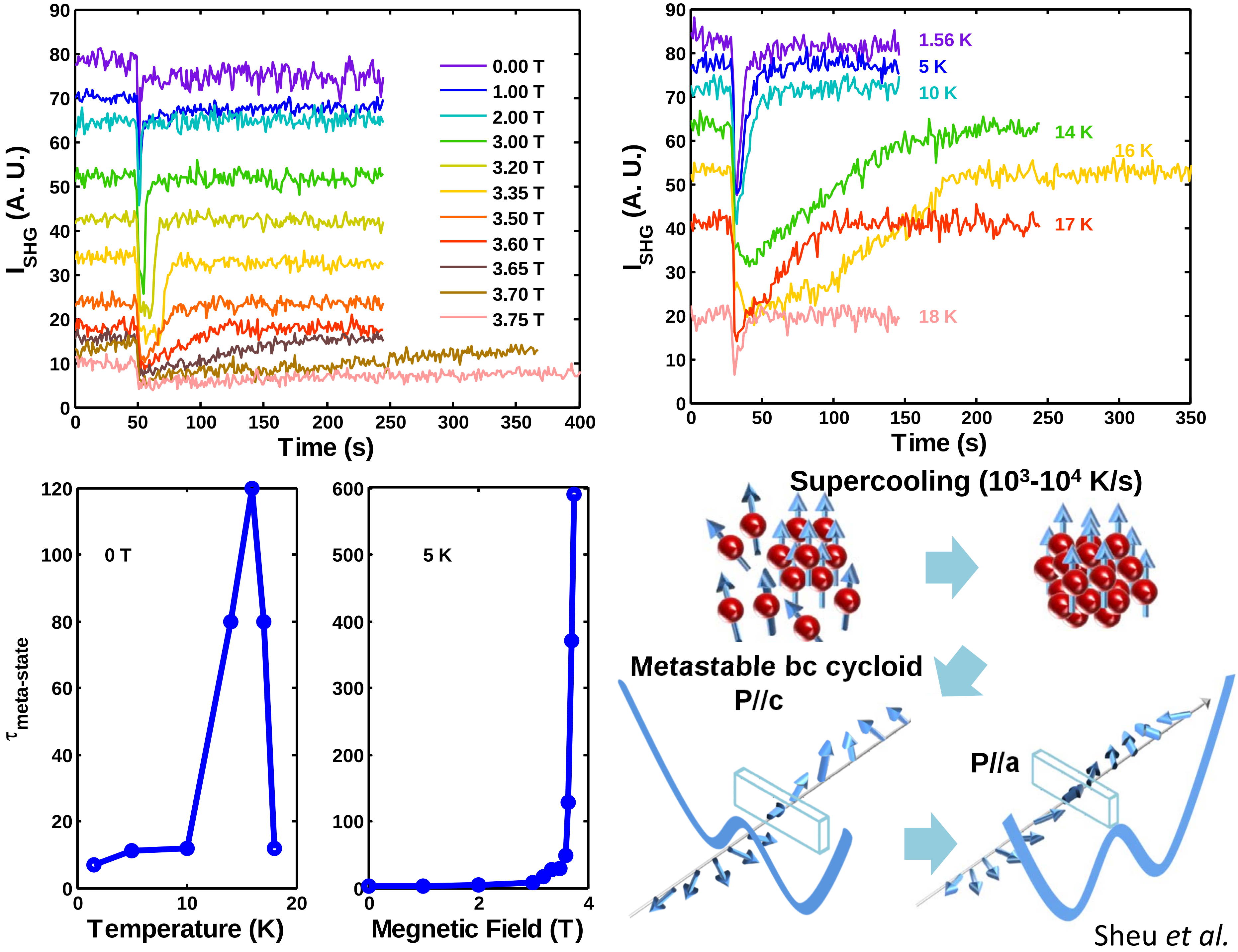

I_{SHG} (A. U.)
Time (s)
0.00 T
1.00 T
2.00 T
3.00 T
3.20 T
3.35 T
3.50 T
3.60 T
3.65 T
3.70 T
3.75 T
1.56 K
5 K
10 K
14 K
16 K
17 K
18 K
$\tau_{meta\text{-}state}$
0 T
Temperature (K)
5 K
Megnetic Field (T)
Supercooling (10^3-10^4 K/s)
Metastable bc cycloid
P//c
P//a
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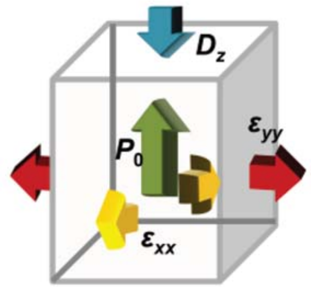
Conclusion

- Demonstration of indirect ME coupling through using ultrashort pulses to excited the FM and probe (proper) FE material composed in a heterostructure.



- Creating a metastable *bc* spiral through intense ultrashort pulse in the *ab* spiral ground state of multiferroic EYMO (improper FE).