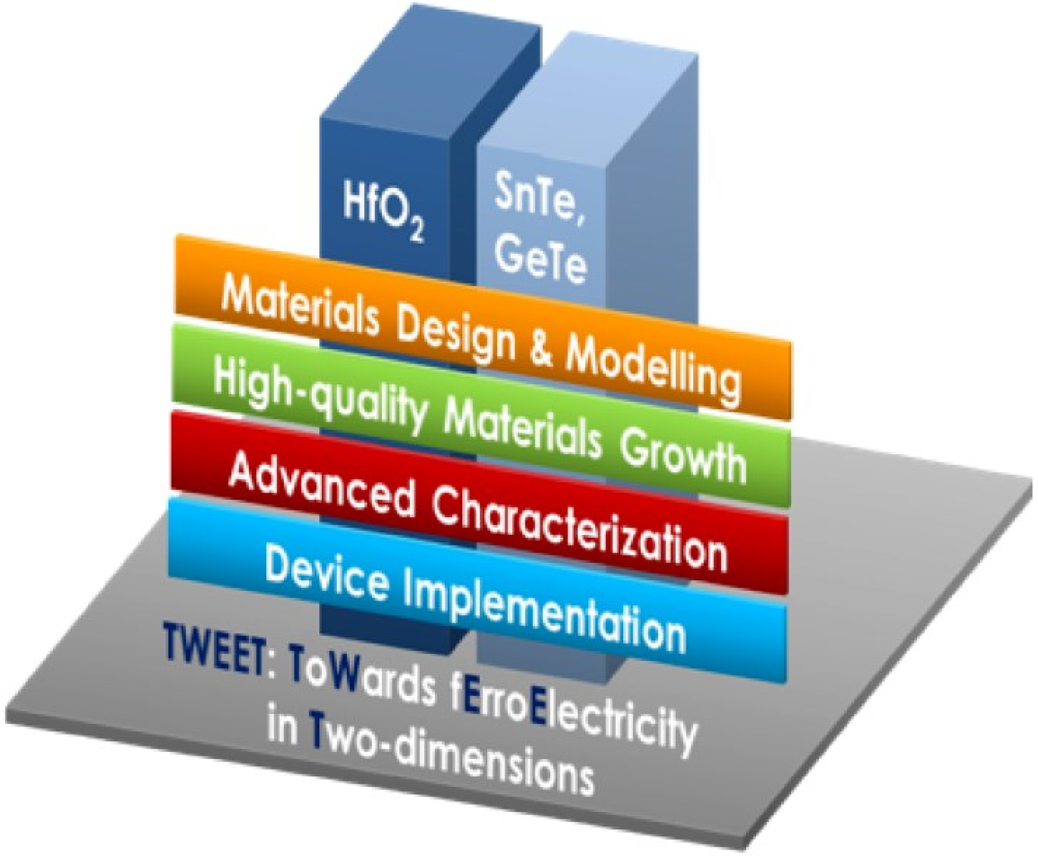
TWEET final Talk

Second harmonic Generation on 2D ferroelectrics



Rohit Kumar
Department of Physics, Universituy of Naples, Federico II.

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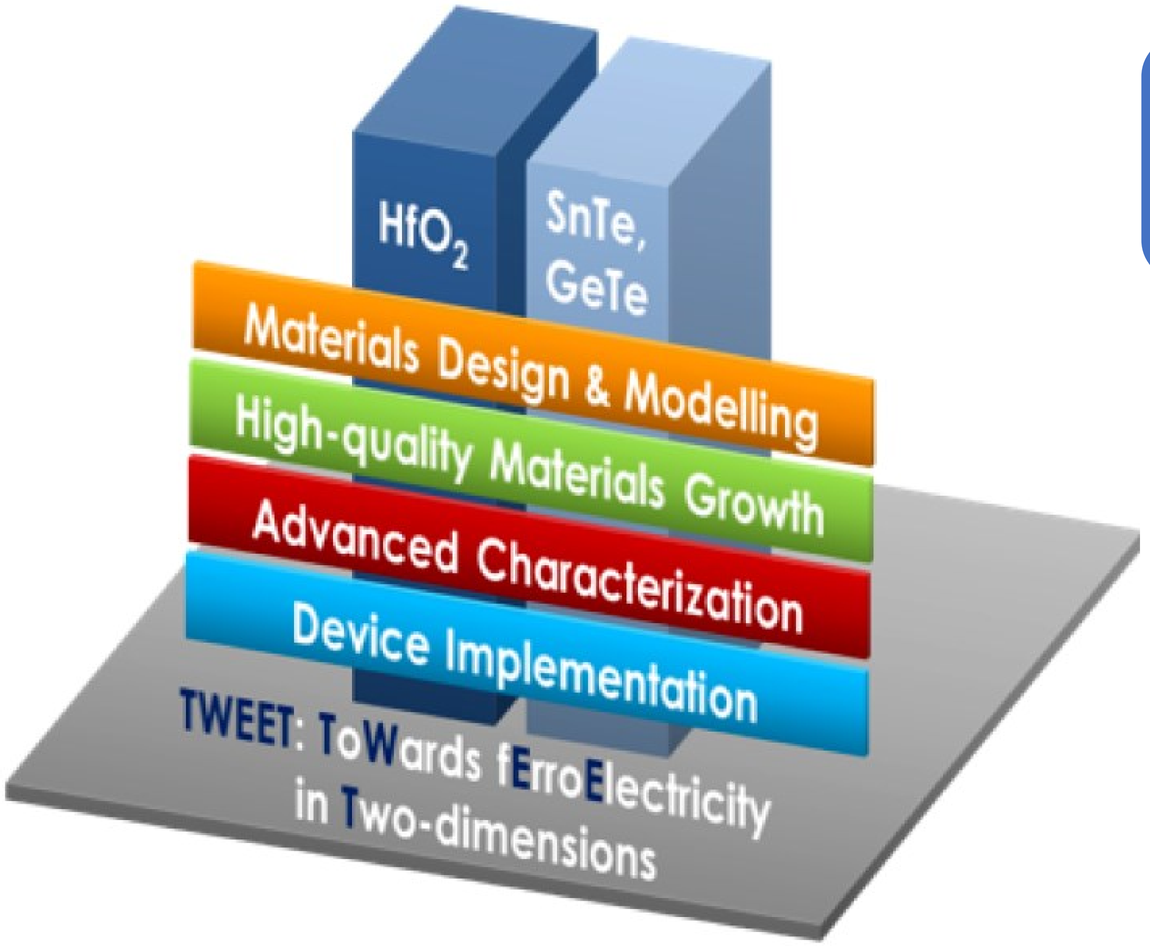
TWEET overview



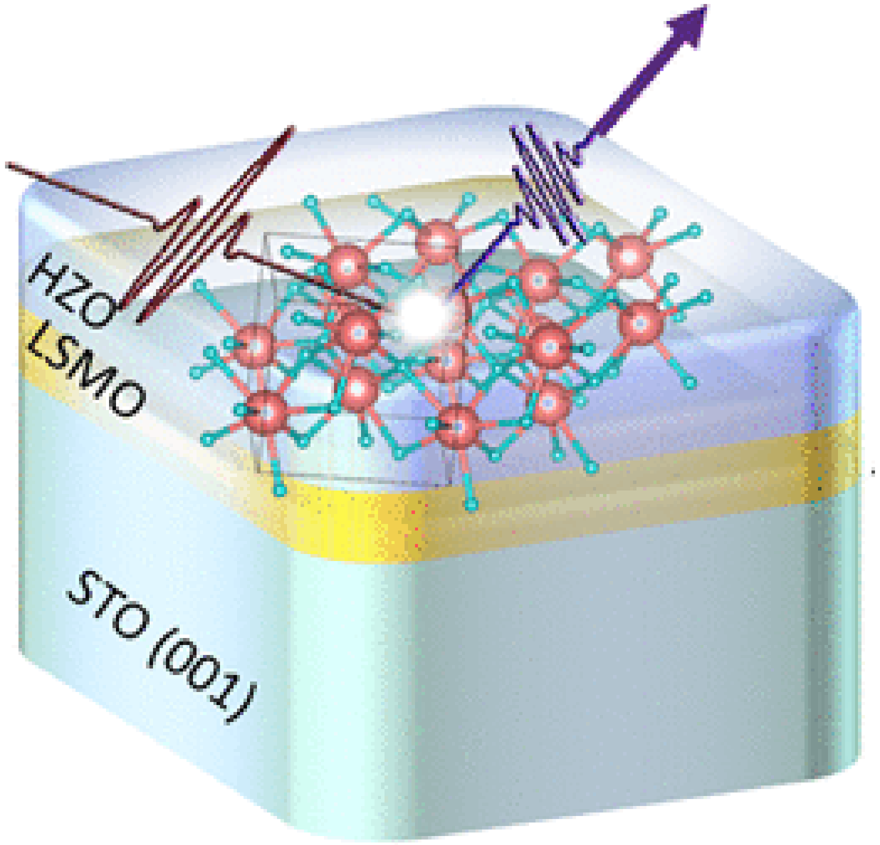
Material Design and modelling

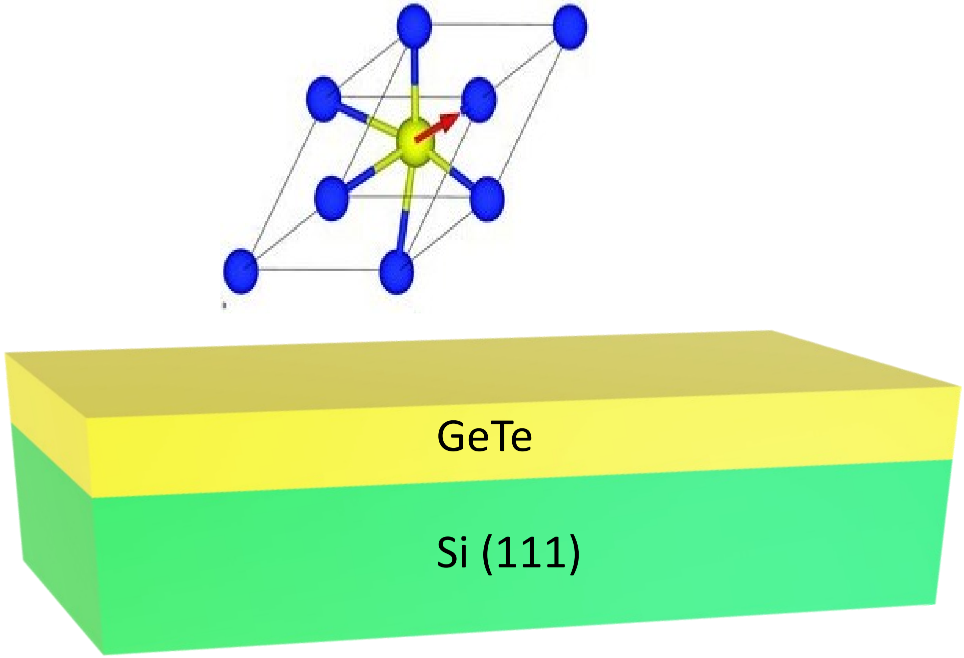
- CNR-SPIN Chieti

Fabrication and standard characterization

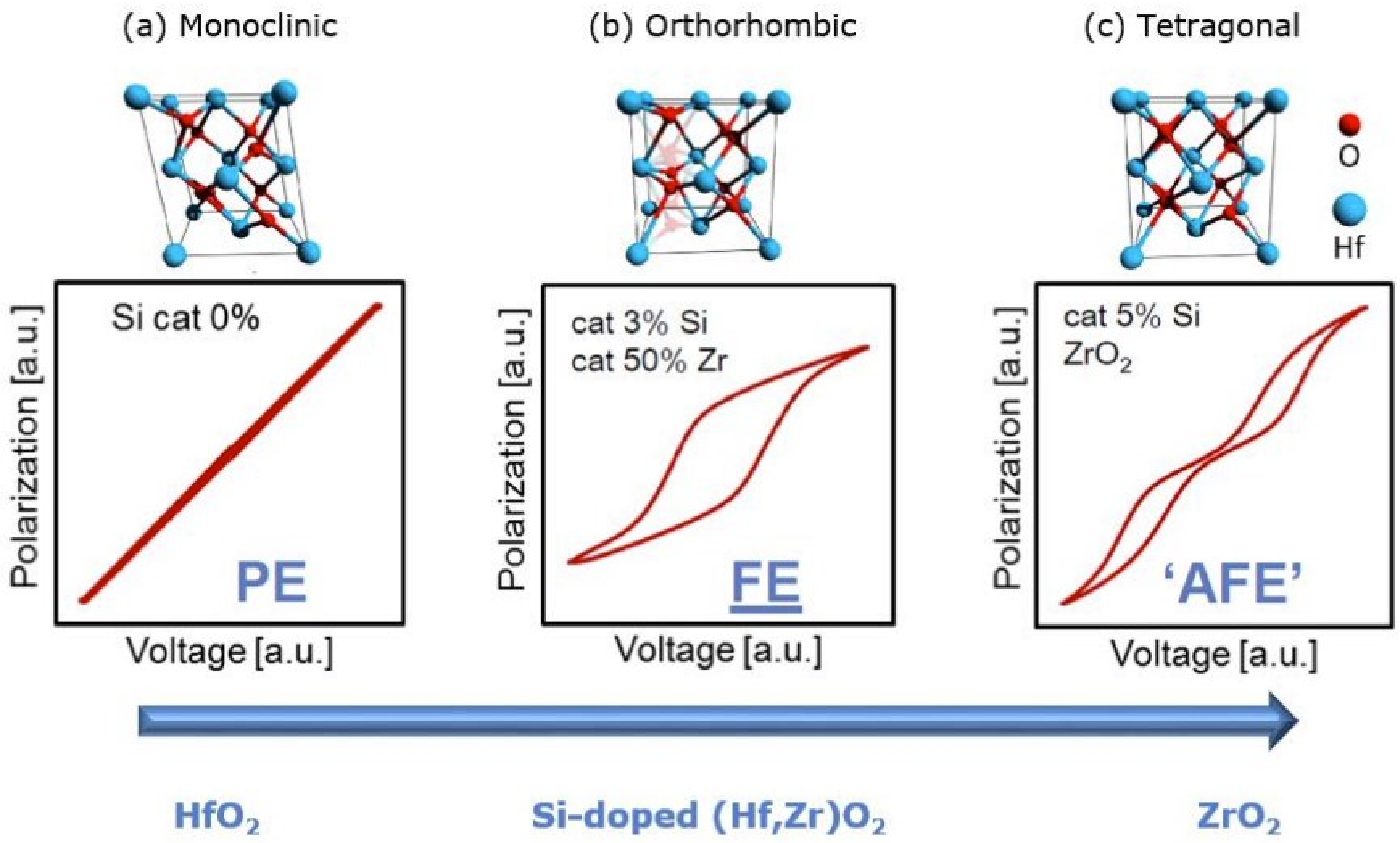
Materials:

Hafnium Zirconium Oxide (HZO)
Germanium Telluride (GeTe)





Hafnium Zirconium Oxide
Crystalline phases of P-E cycle for different compositions of the system

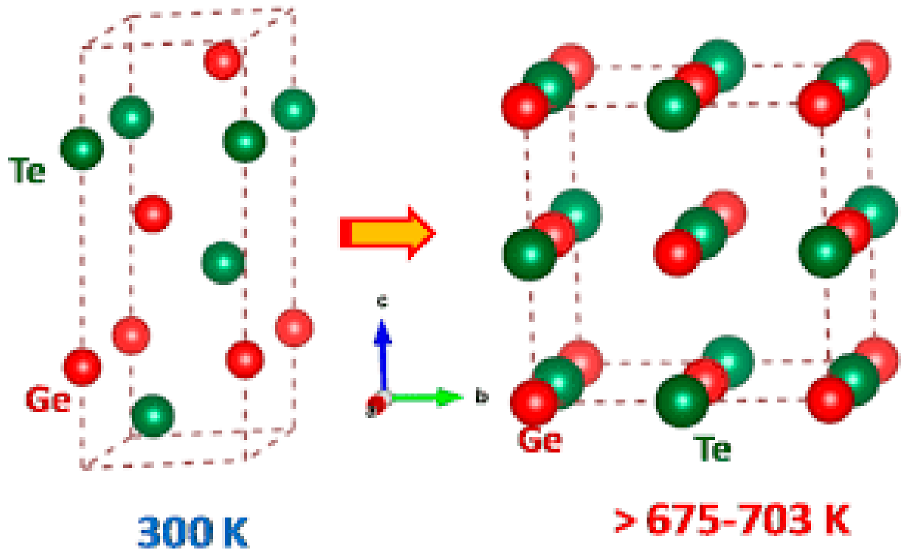


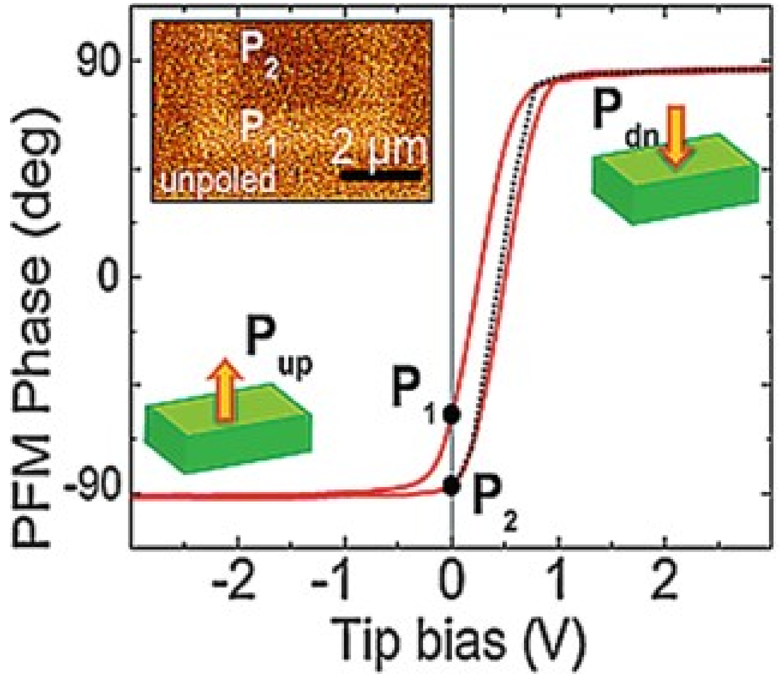
Claudia Richter, et .al. Si Doped Hafnium Oxide—A “Fragile” Ferroelectric System. Adv. Electron.Mater.2017, 3, 1700131.

Germanium Telluride (GeTe)

Germanium Telluride

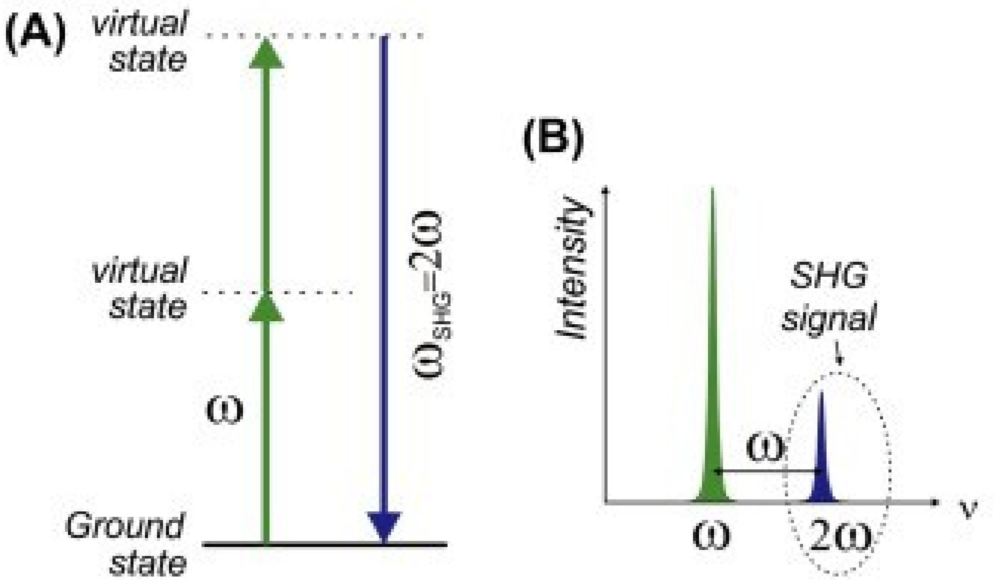
- GeTe is Ferroelectric Rashba Semiconductor
- GeTe is Silicon compatible
- Rhombohedral unit cell of GeTe (polarization pointing along the pseudo-cubic [111] axis)
- FE hysteresis loop of GeTe(111) measured by piezo-force microscopy





Liebmann M et.al. Adv. Mat. (2016) 28:560–565

Second Harmonic Generation (SHG)



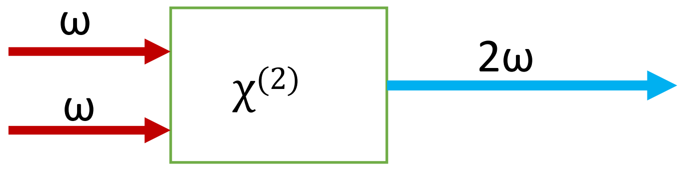
Laser matter interaction

$$P = \epsilon_0(\chi^{(1)}E_\omega + \chi^{(2)}E_\omega^2 + \chi^{(3)}E_\omega^3 + \cdots)$$
$$= P^{(1)} + P^{NL}$$

$\chi^{(1)}$ describes the linear optics, e.g., how lens work.

$\chi^{(2)}$ describes the second order effects such as SHG.

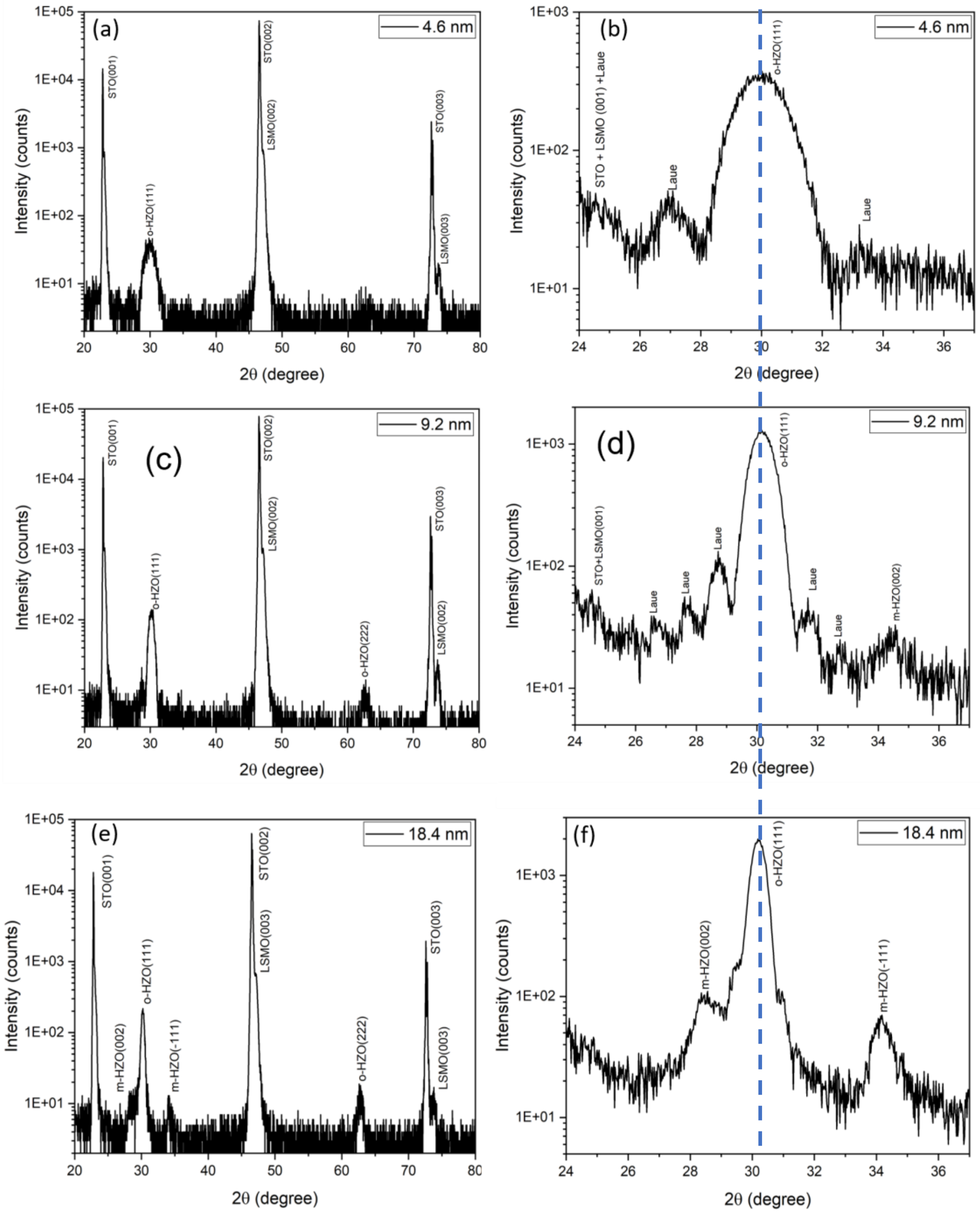
Ferroelectrics



Electric dipole SHG $P_i(2\omega) = \chi^{(2)}_{ijk} E_j(\omega)E_k(\omega)$

Electric Quadrupole SHG $Q_i(2\omega) = \chi^{(2)}_{ijkl} E_j(\omega)K_kE_l(\omega)$

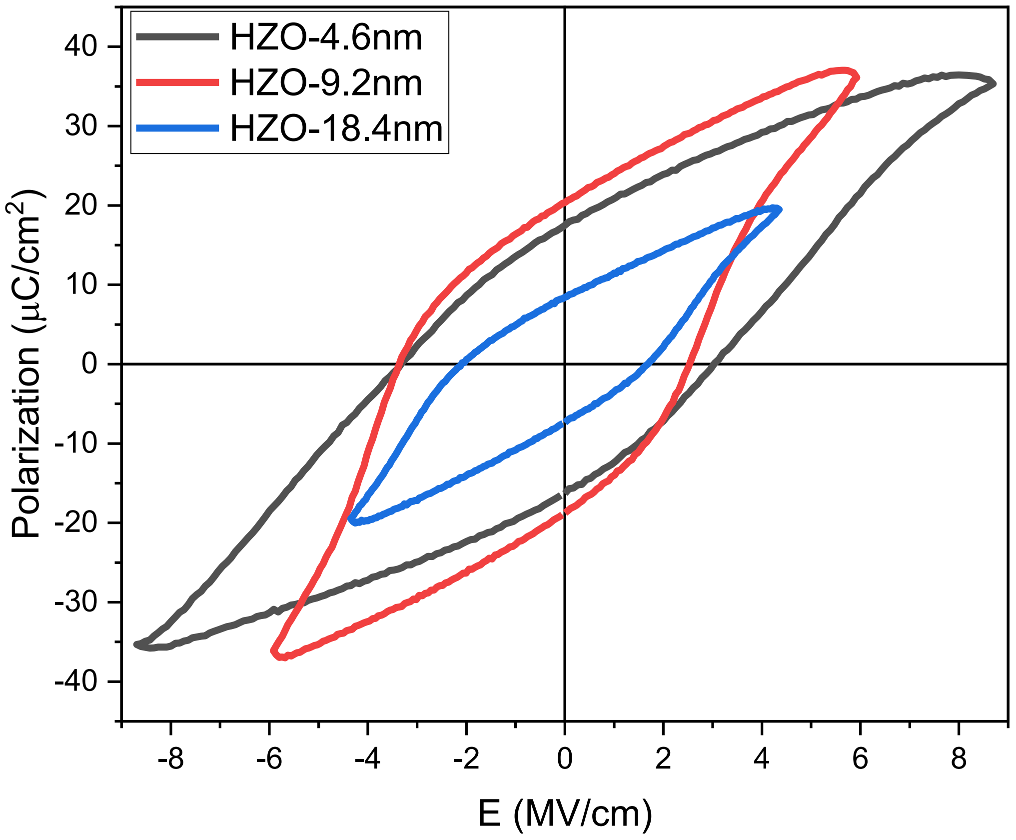
XRD Results of HZO



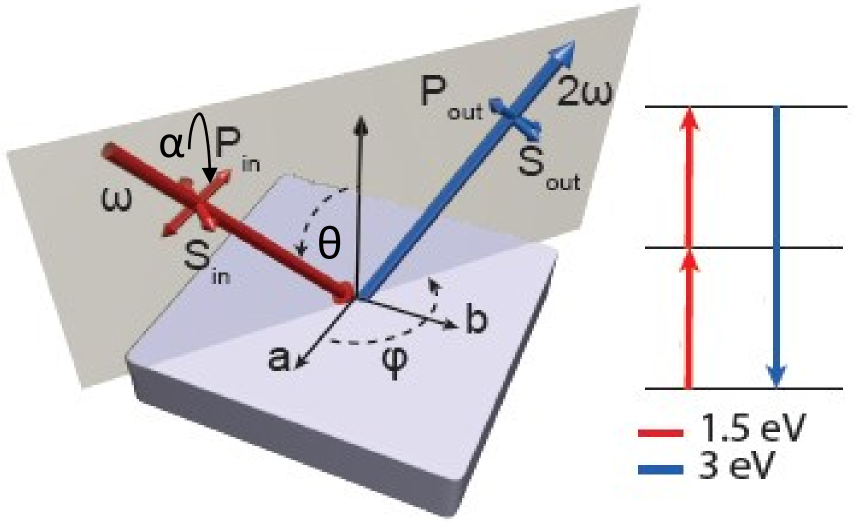
- Peak shifts at smaller angles indicating d_{111} elongation in thinner films (orthorhombic phase)
- 9.2 nm HZO thin film has largest remanent polarization

Done at ICMAB

Electrical measurements

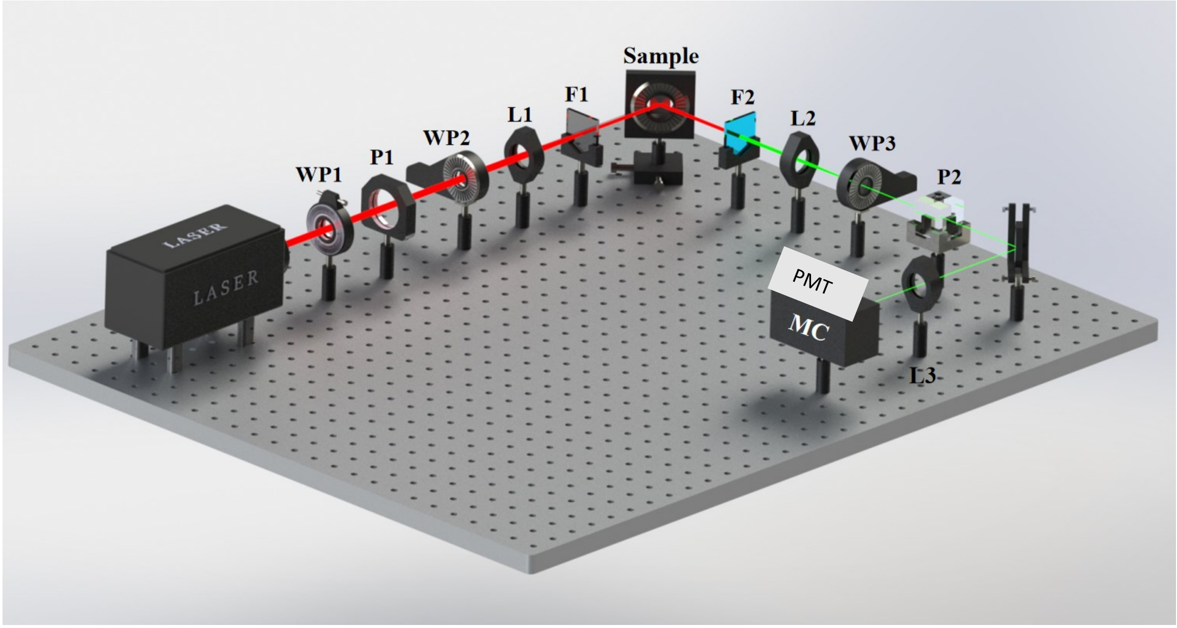


Experimental Geometry

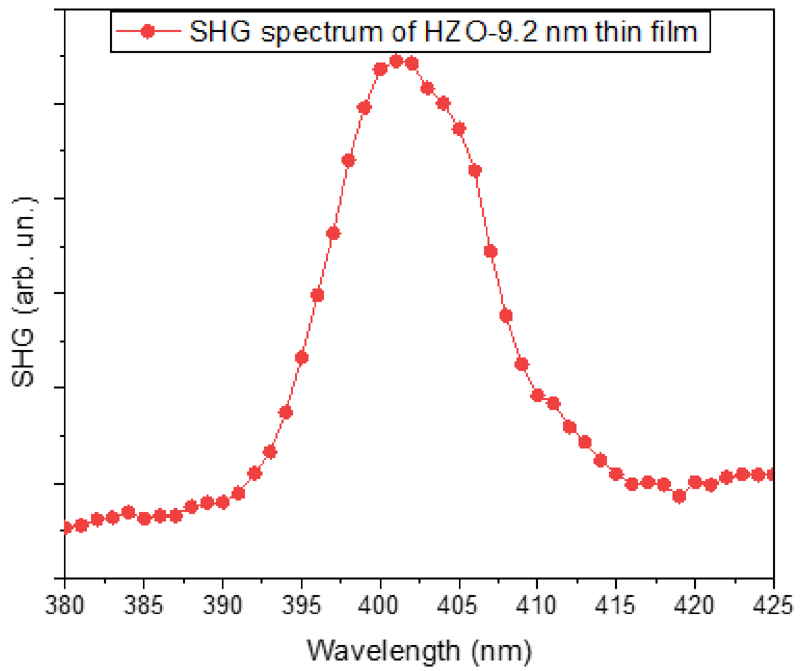


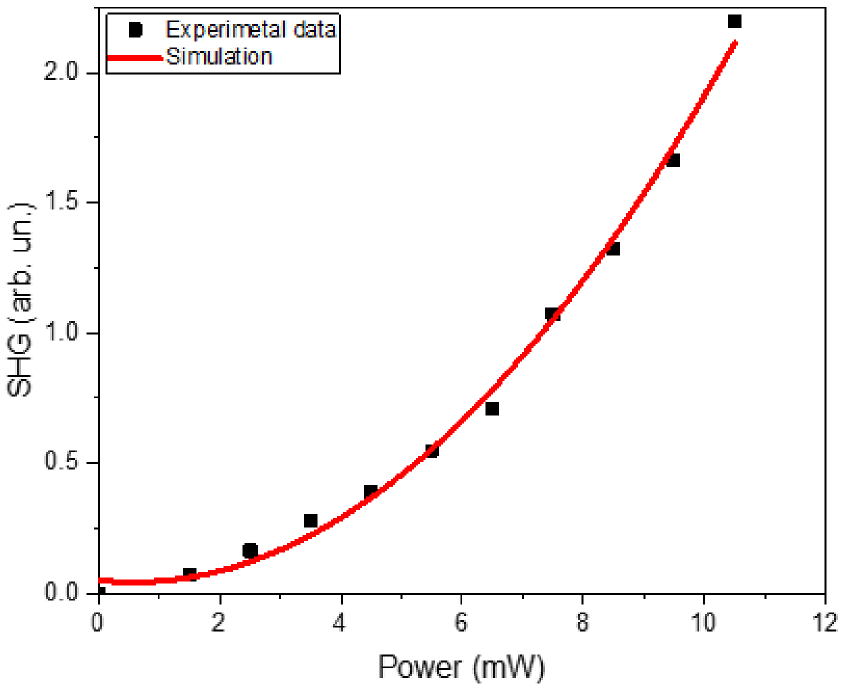
Alpha α : Polarization angle
ϕ : Sample azimuthal angle
θ : Angle of incidence

Experimental set up (Beam path)



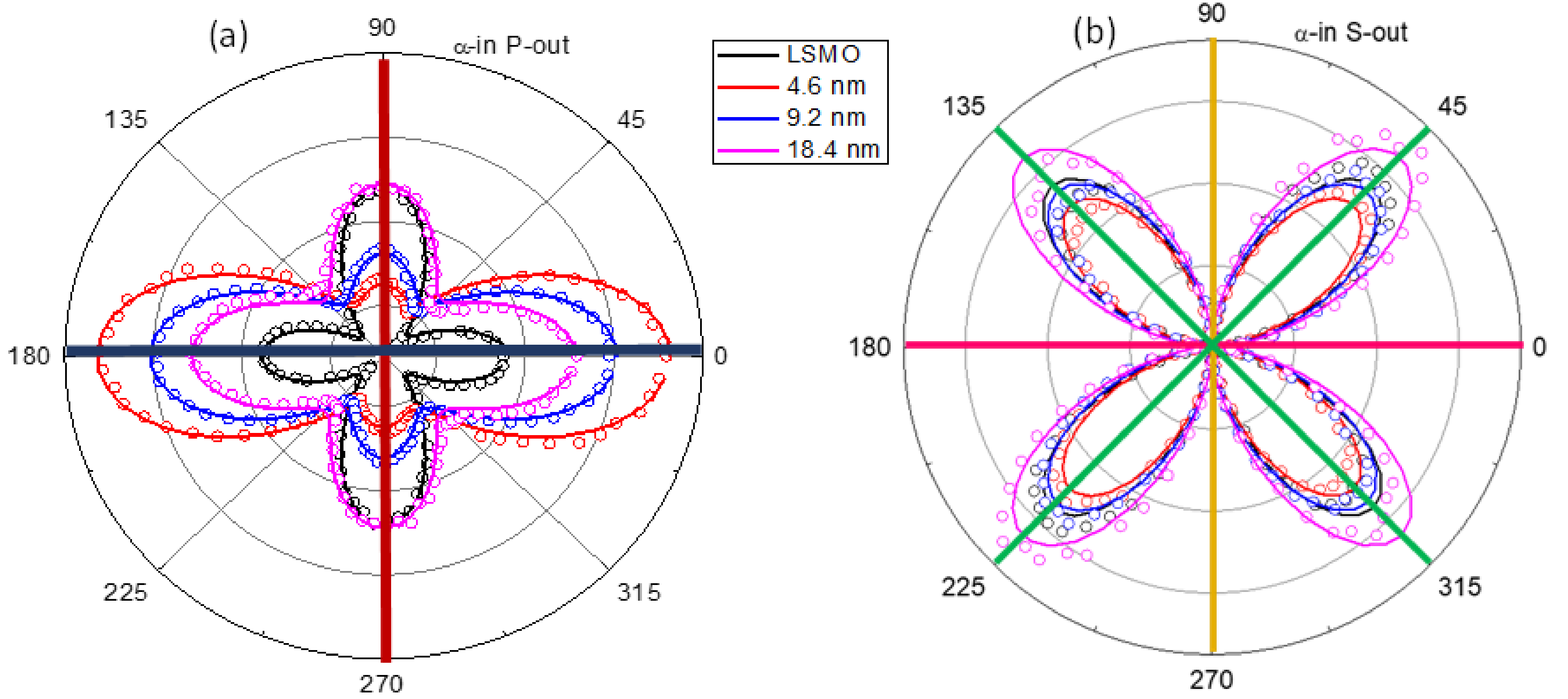
Spectral and power characterization of SHG



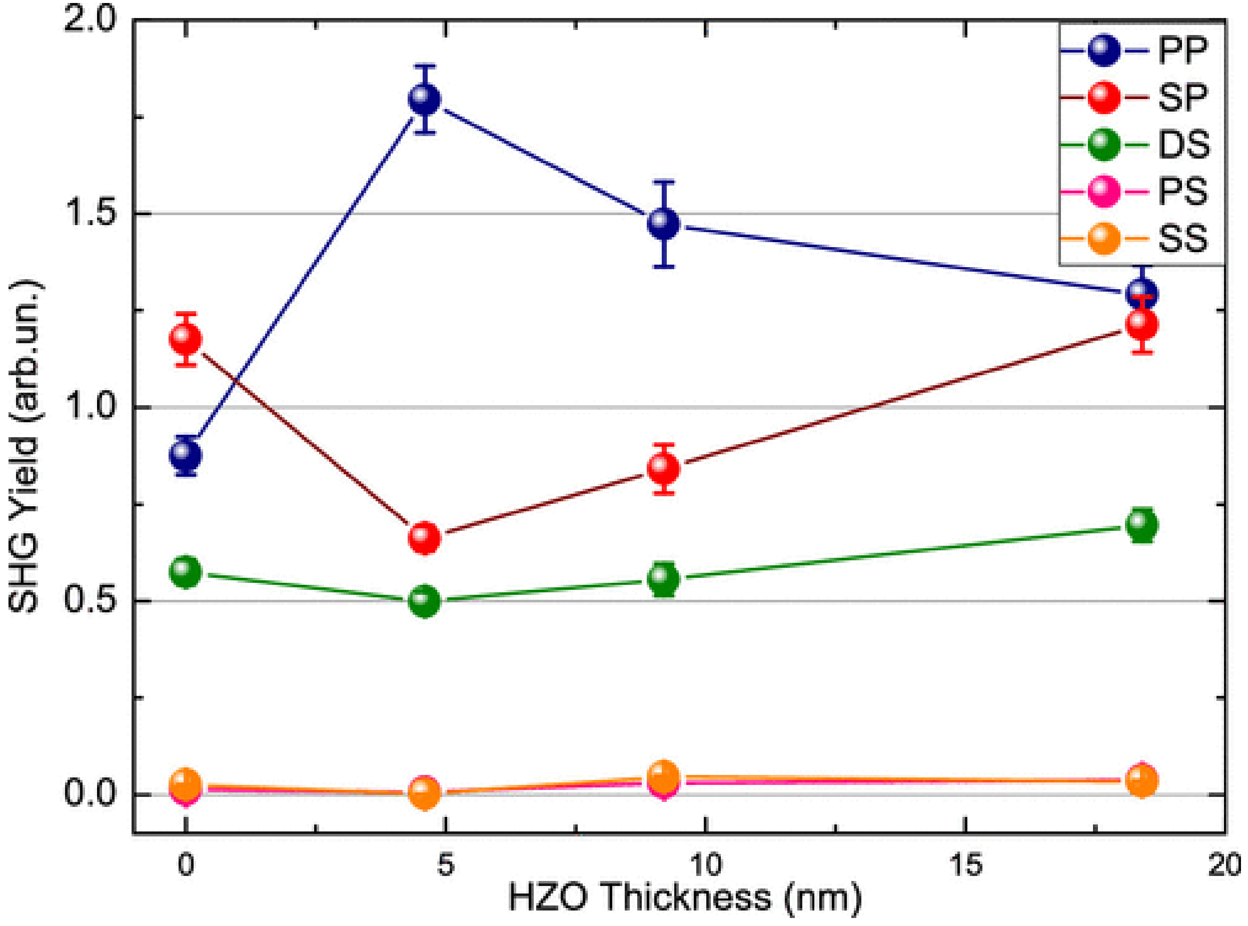


SHG results

Alpha scans

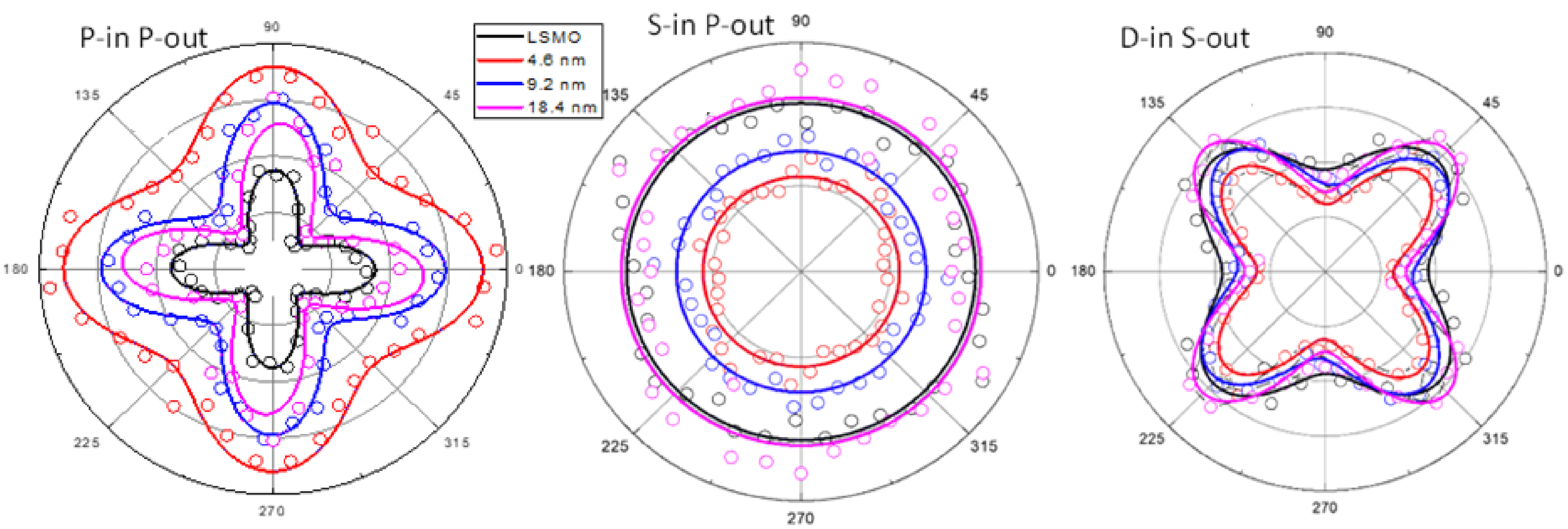


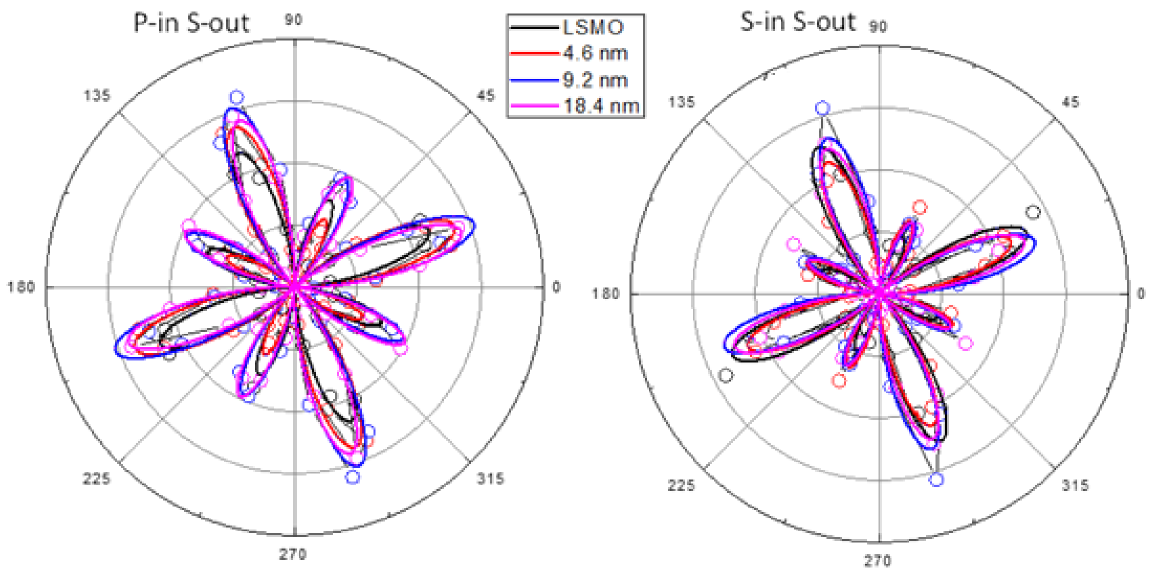
SHG signal in different polarization combination



- HZO coverage affects the SHG components
- PP has a maximum for thinnest and SP has a minimum

Azimuthal Scans

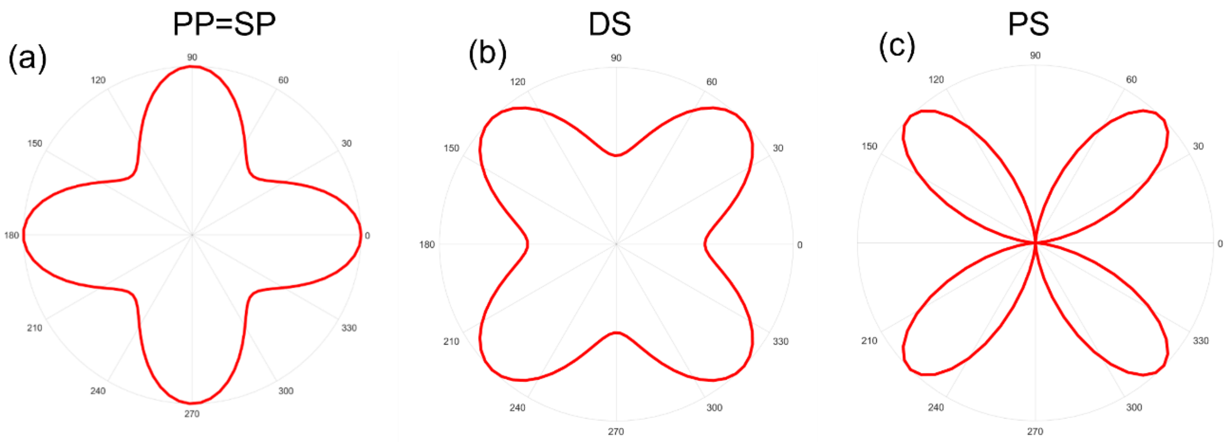




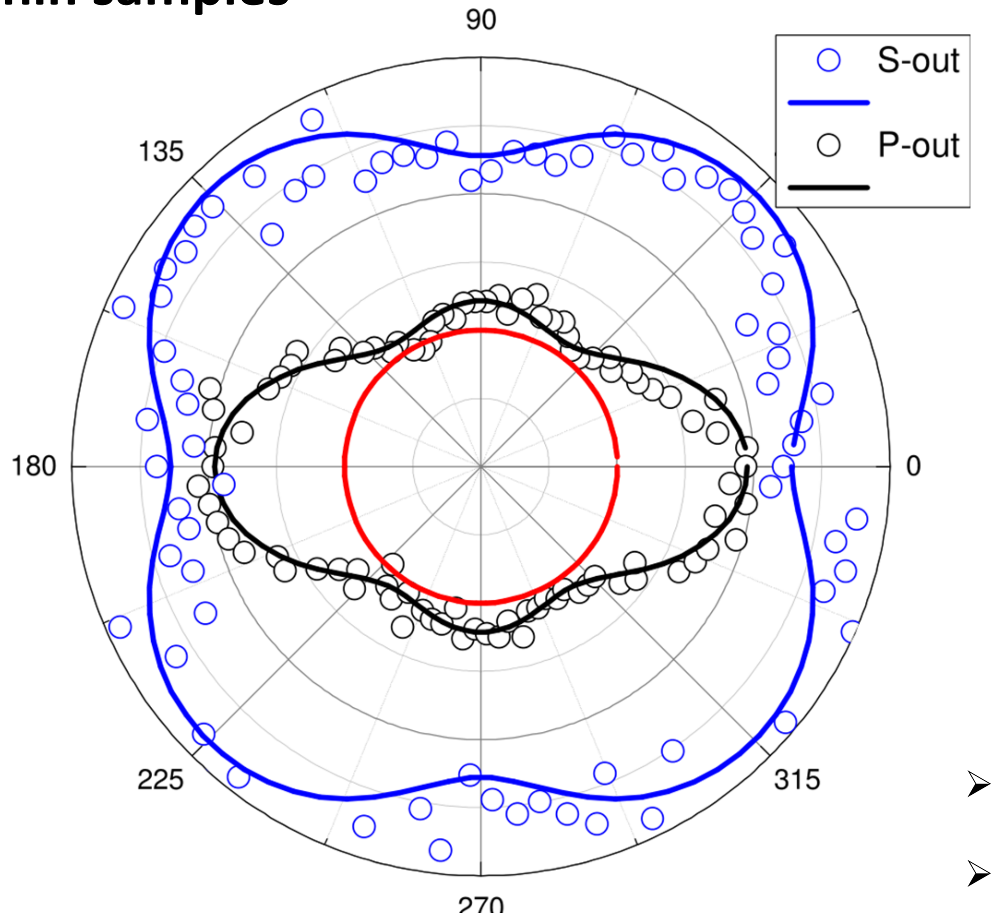
- Five non-vanishing polarization combinations but they don't belong to the same origin
- PS and SS are not changing with the film thickness
- No single point group symmetry that can fit the data

Looking for a real symmetry ?

Symmetry	Non-vanishing elements
o-HZO, mm2	xzx = xxz yyz = yzy zxx zyy zzz
m-HZO, 2/m	Each element vanishes.
t-HZO, 4/mmm	Each element vanishes.



Ratio b/w LSMO-thick and LSMO-thin samples



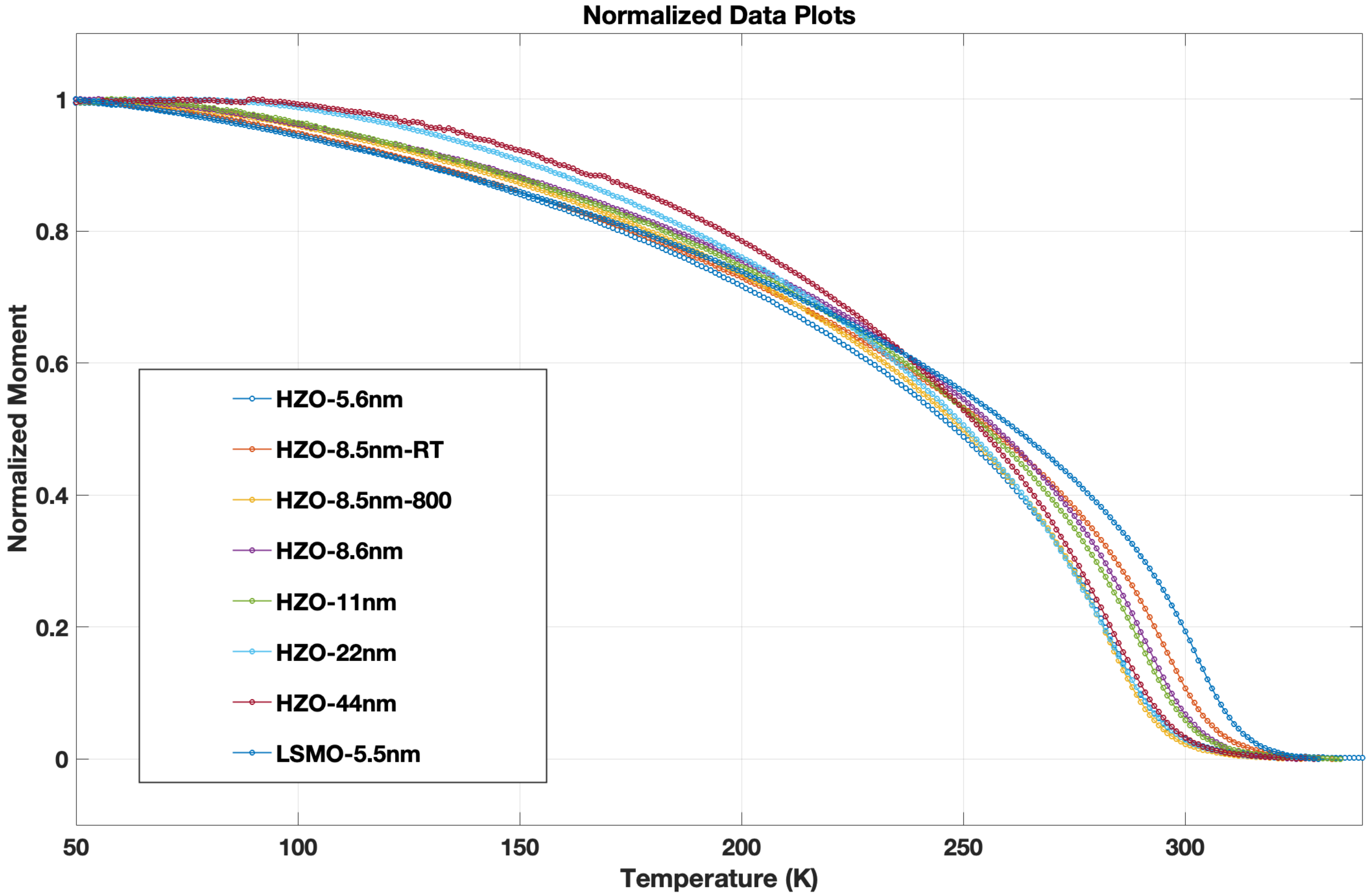
Blue curve represents the S-out ratio between LSMO thick and LSMO thin samples.

Set 2: LSMO = 8 and 20 nm
HZO = 9 nm

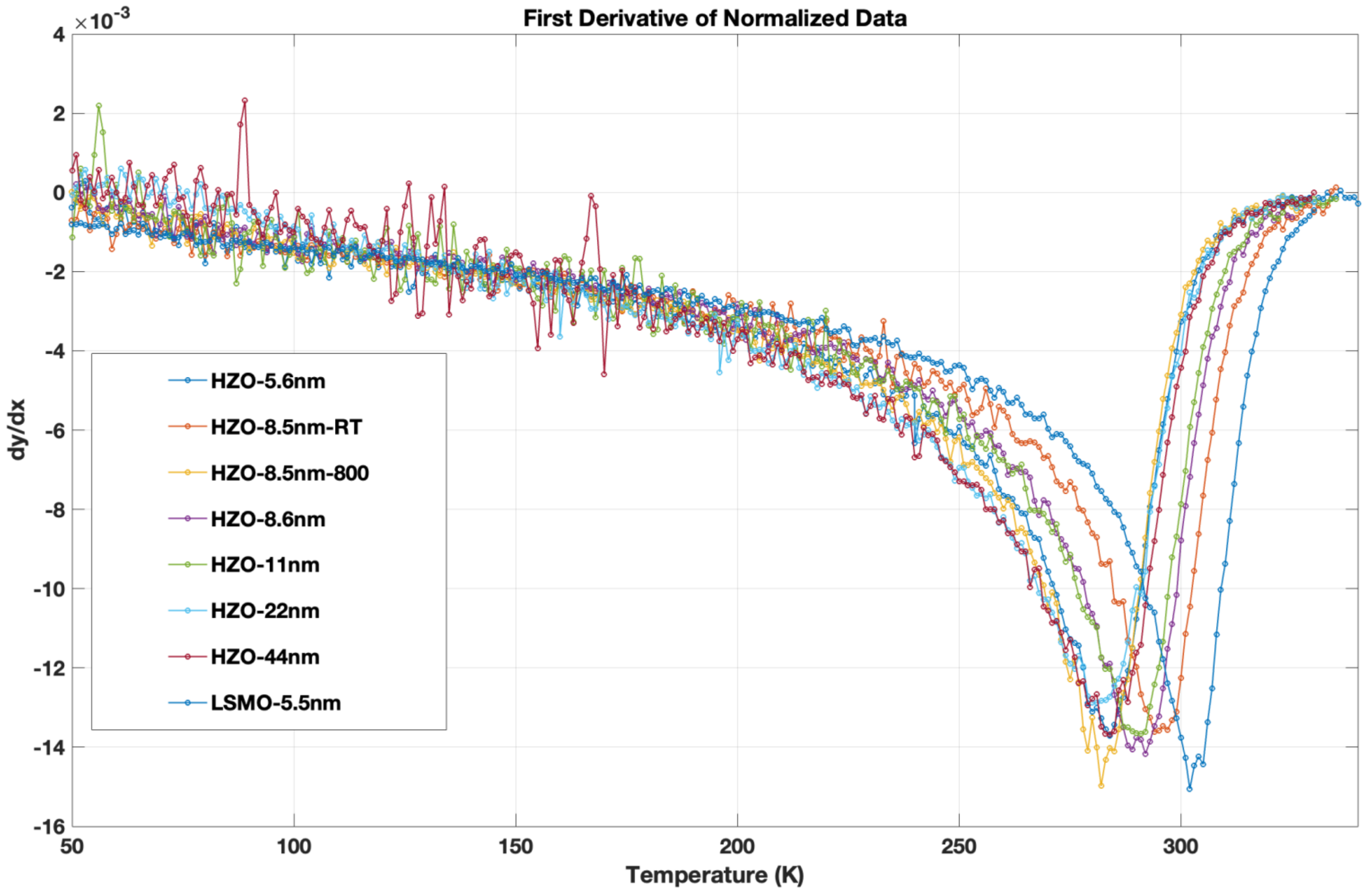
- If the SHG signal was scaling with LMSO then we should have a circle.
- In case of S-out, it seems a circle but in P-out, there is an elongation in PP direction and SP component is lying almost around 1.
- This suggests that SHG cannot be solely LSMO bulk dependent but there is an interfacial contribution too.

Magnetic measurements on HZO thin films

M vs T for all samples

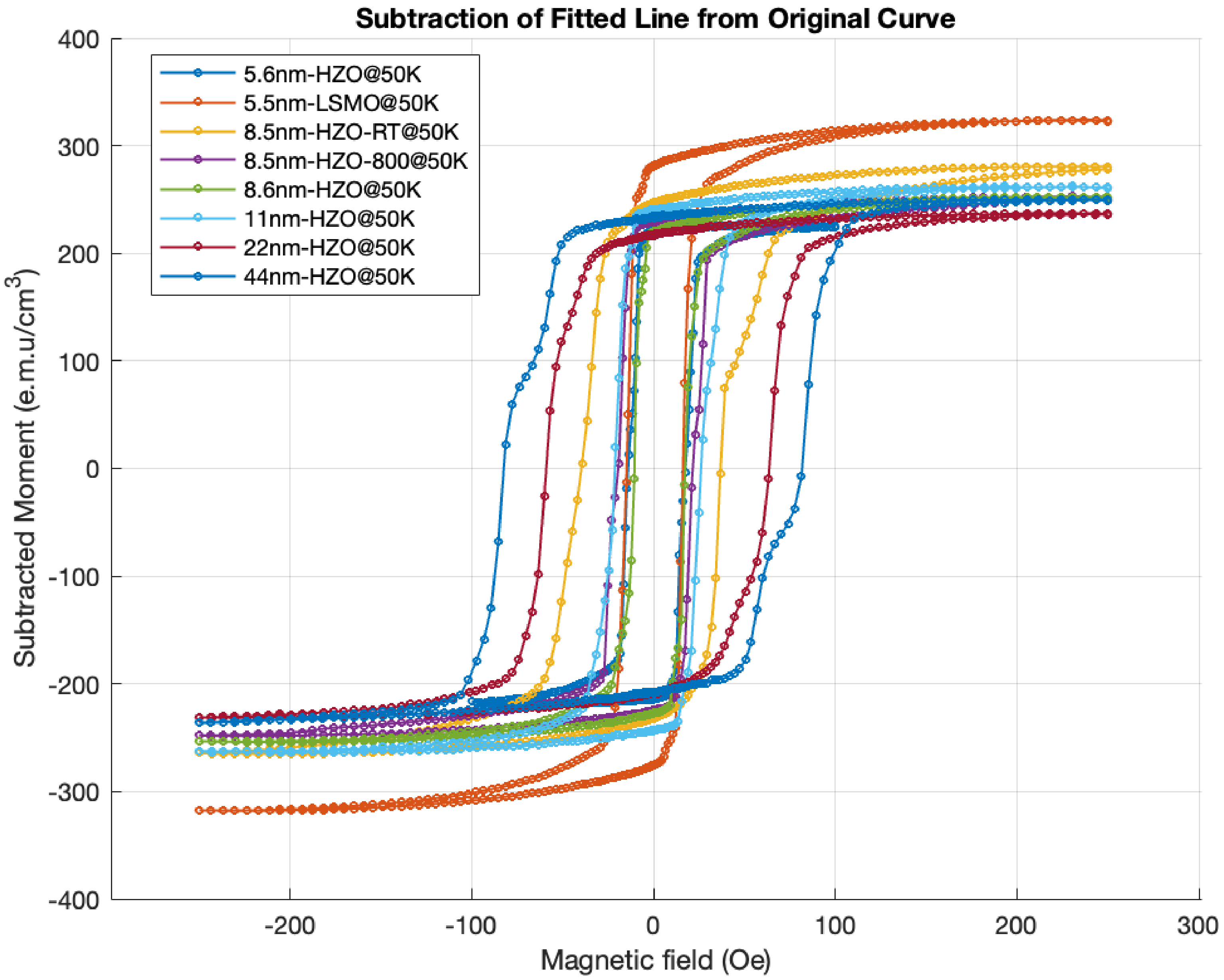


To find Tc, we use first derivative

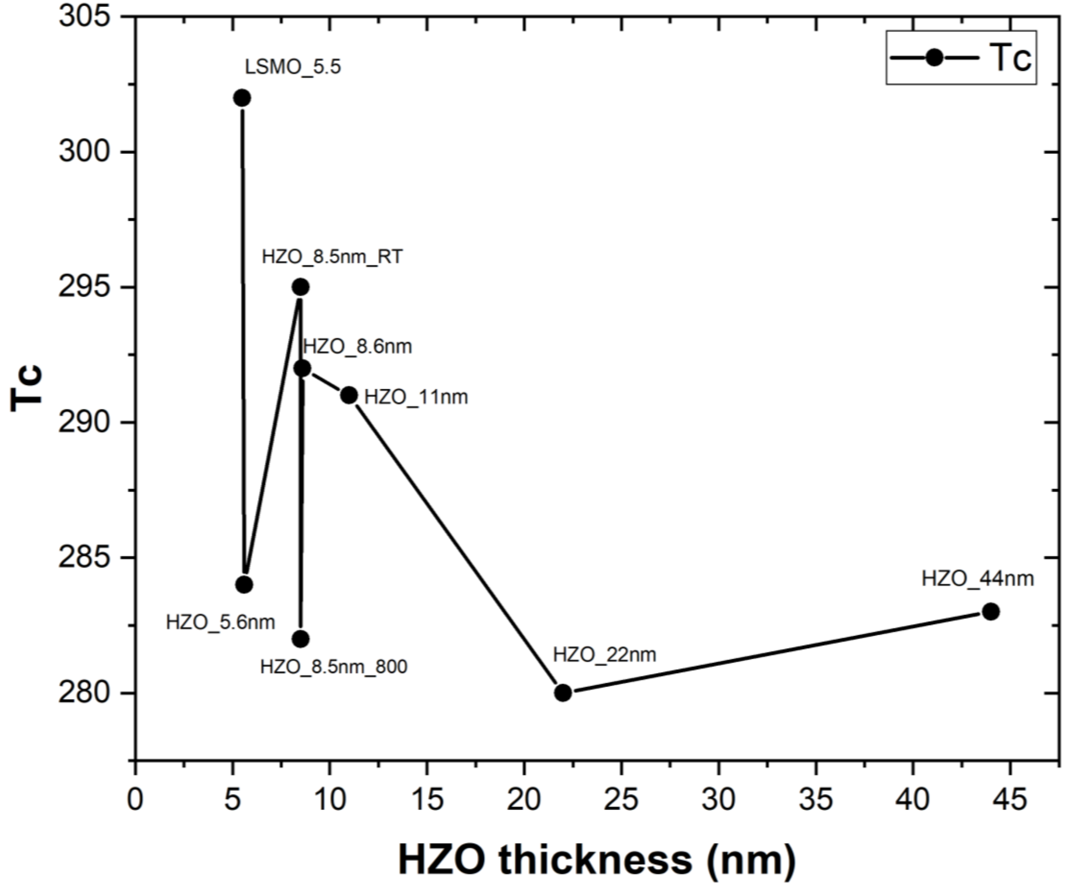


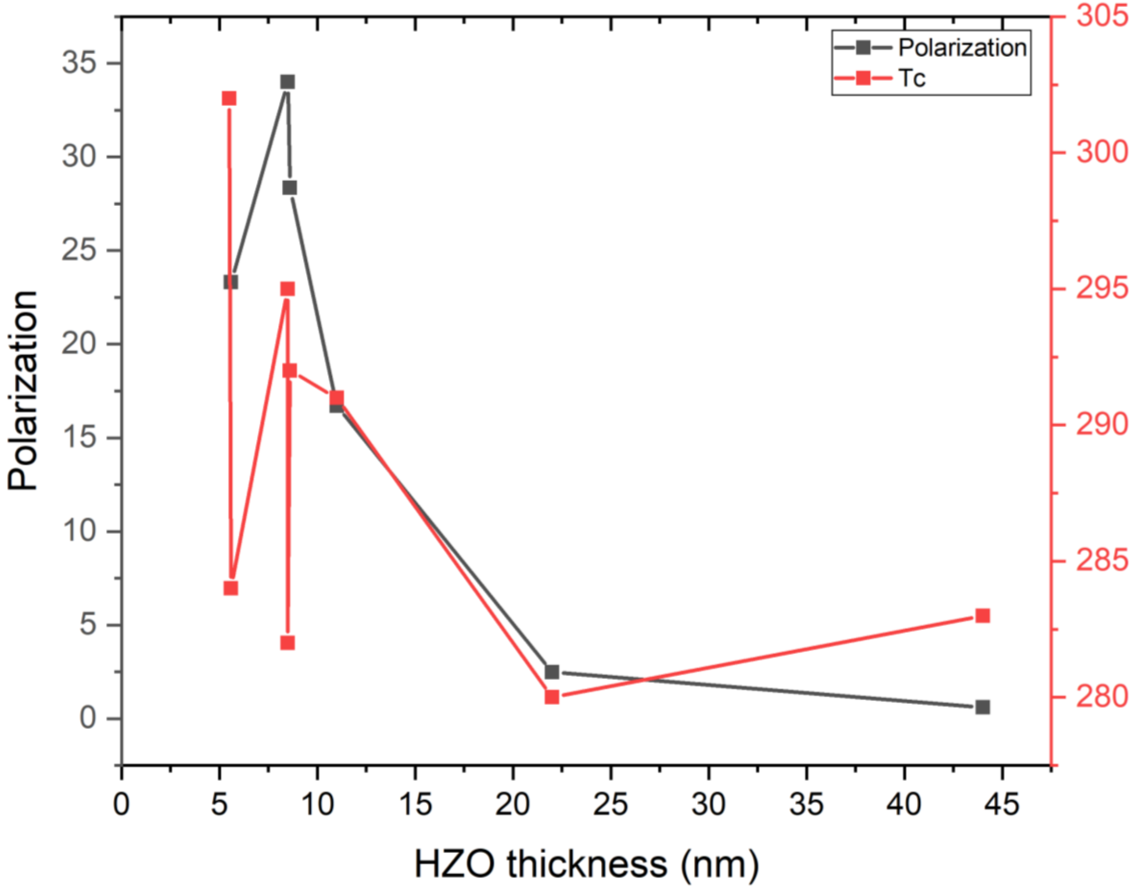
Sample	Tc (K)
LSMO_5.5nm	302
HZO_5.6nm	284
HZO_8.5nm_RT	295
HZO_8.5nm_800	282
HZO_8.6nm	292
HZO_11nm	291
HZO_22nm	280
HZO_44nm	283

M vs H for HZO thin films

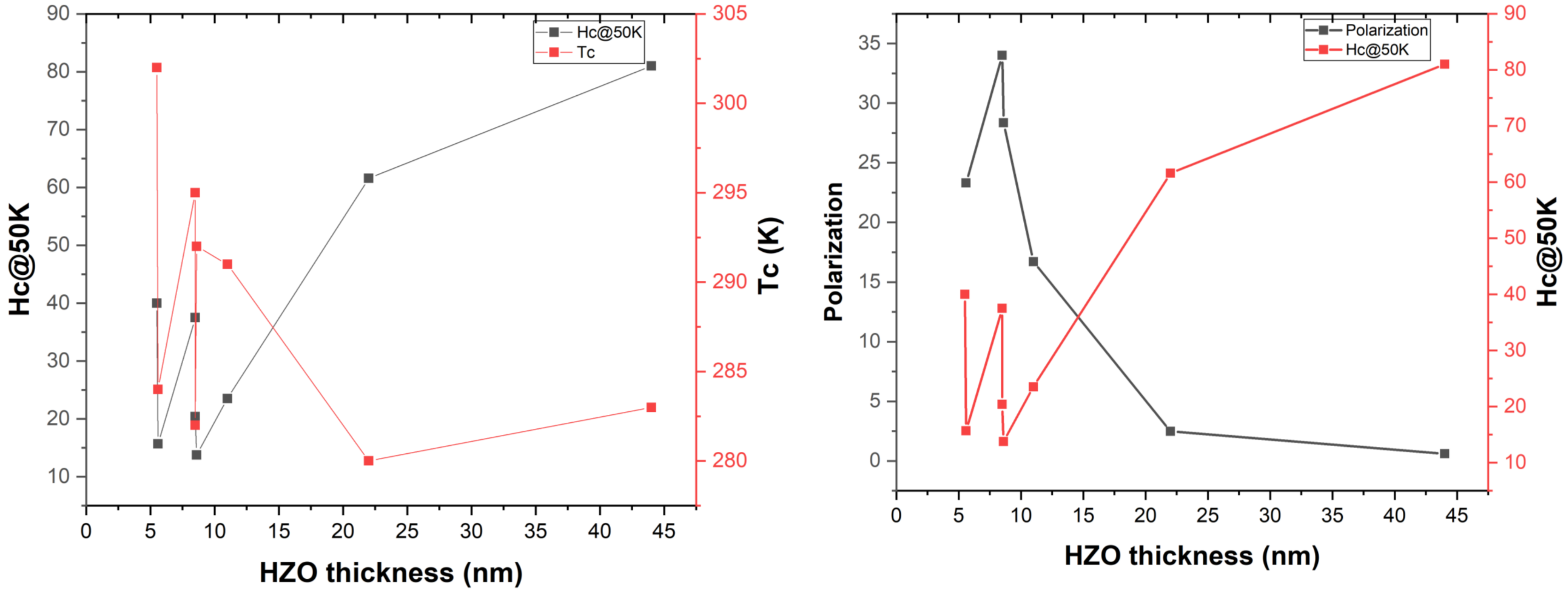


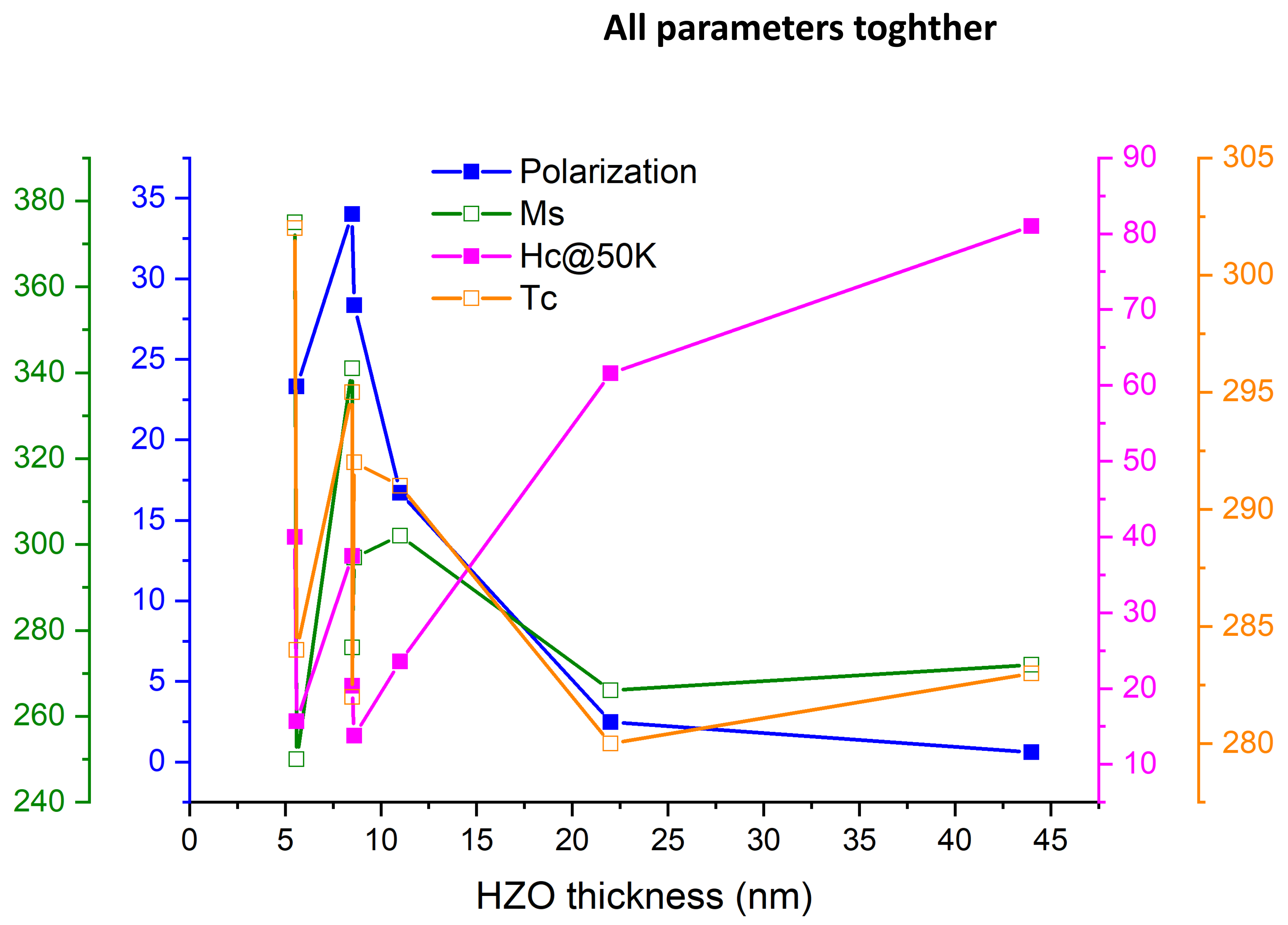
HZO thickness vs Tc vs Polarization





Magnetic coerecivity vs Tc vs Polarization

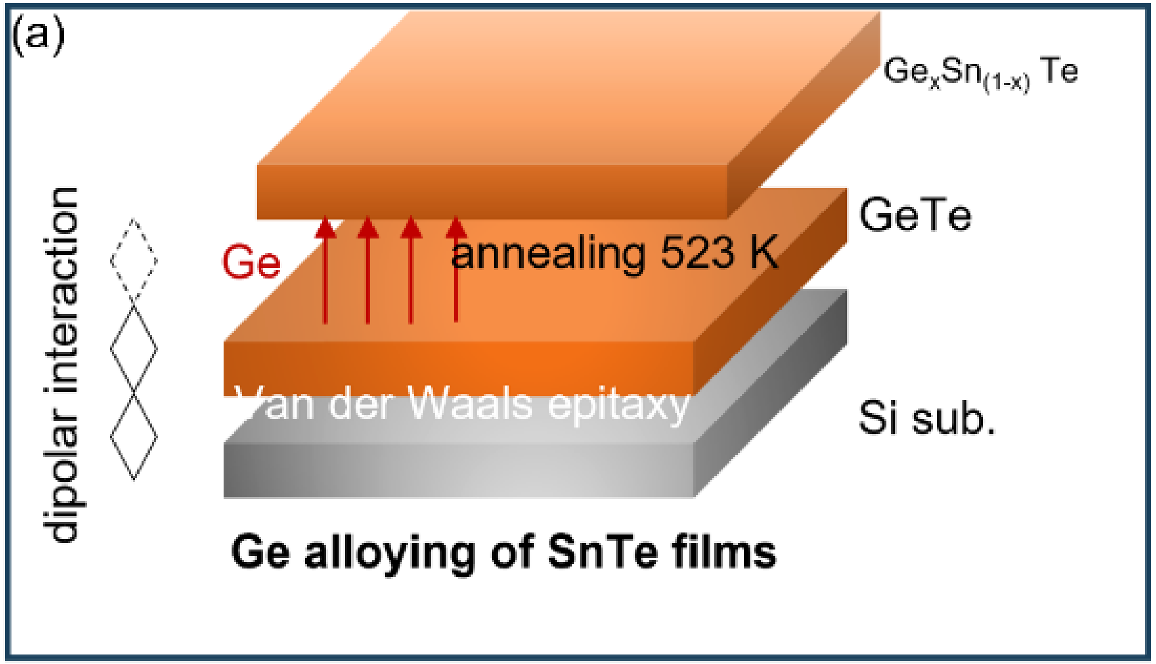


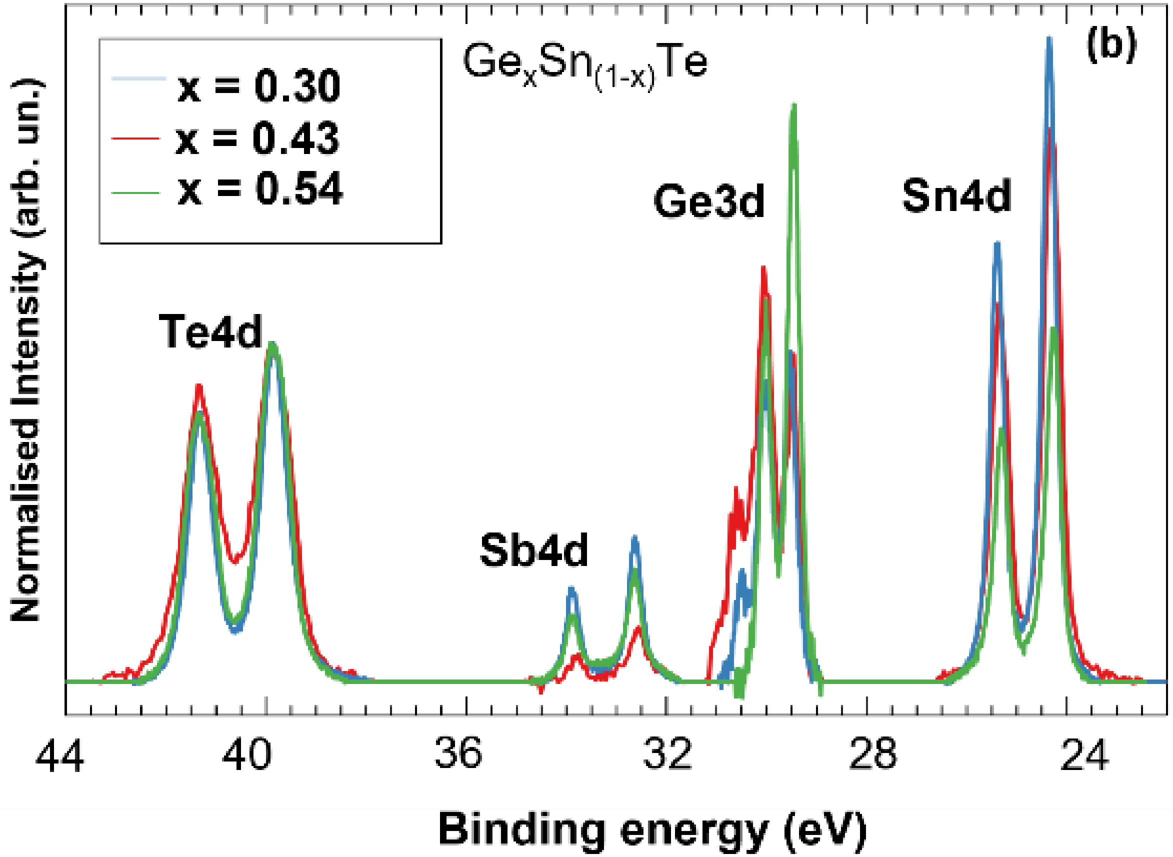


All parameters toghther
Polarization
Ms
Hc@50K
Tc
HZO thickness (nm)
0
5
10
15
20
25
30
35
40
45
240
260
280
300
320
340
360
380
0
5
10
15
20
25
30
35
10
20
30
40
50
60
70
80
90
280
285
290
295
300
305

Part II Germanium Telluride

Growth and Characterization of Pure and doped GeTe

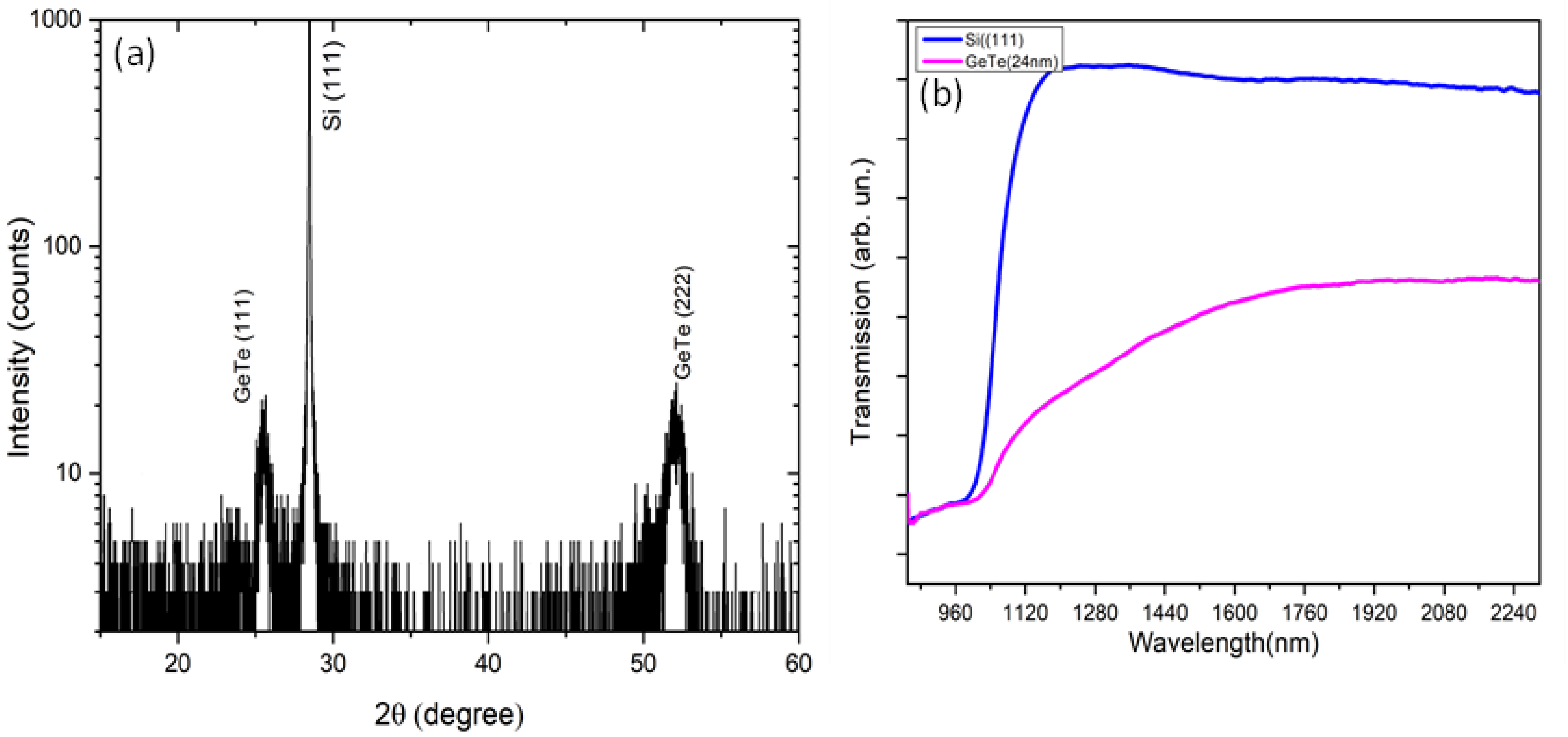




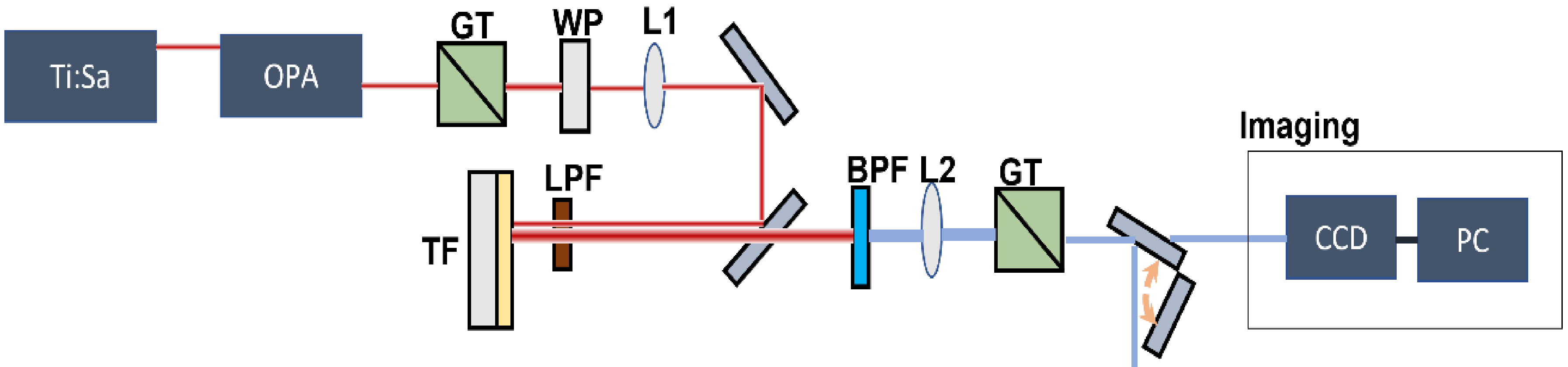
MBE Process: Diffusion of Ge into SnTe matrix when two layers of GeTe and SnTe were grown on top of each other

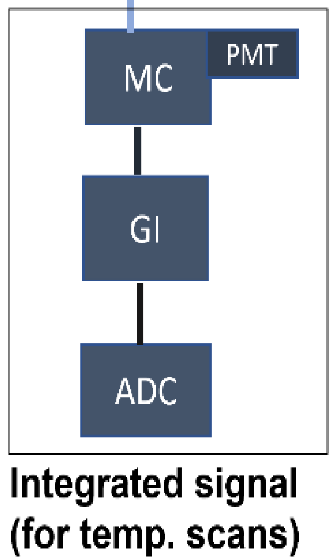
Done by collaboration with PoliMi

XRD and Linear spectra



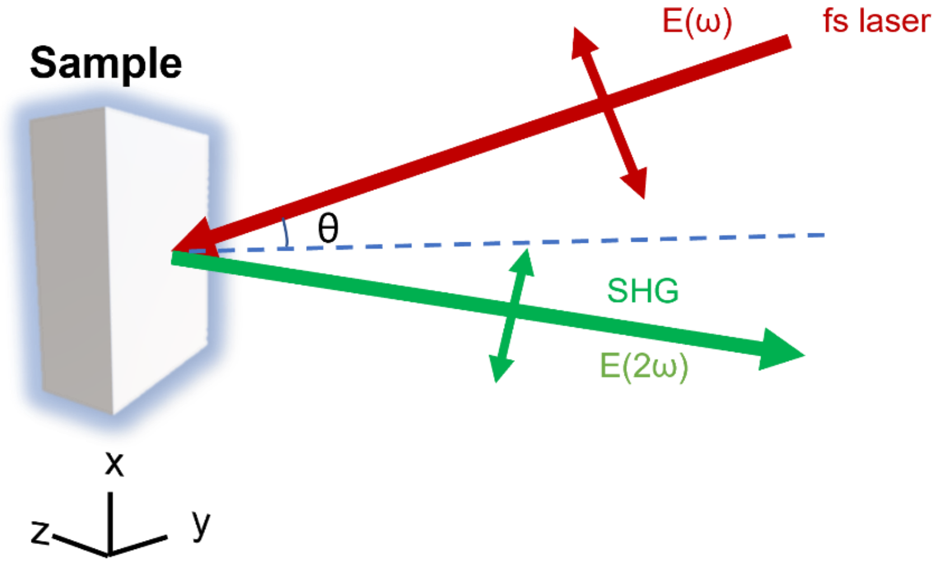
Optical SHG set-up

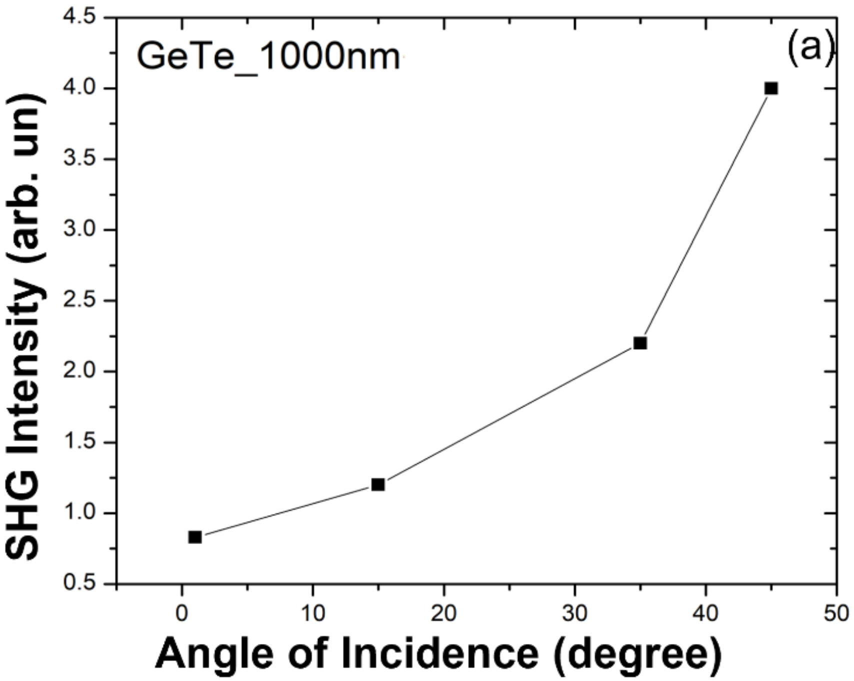


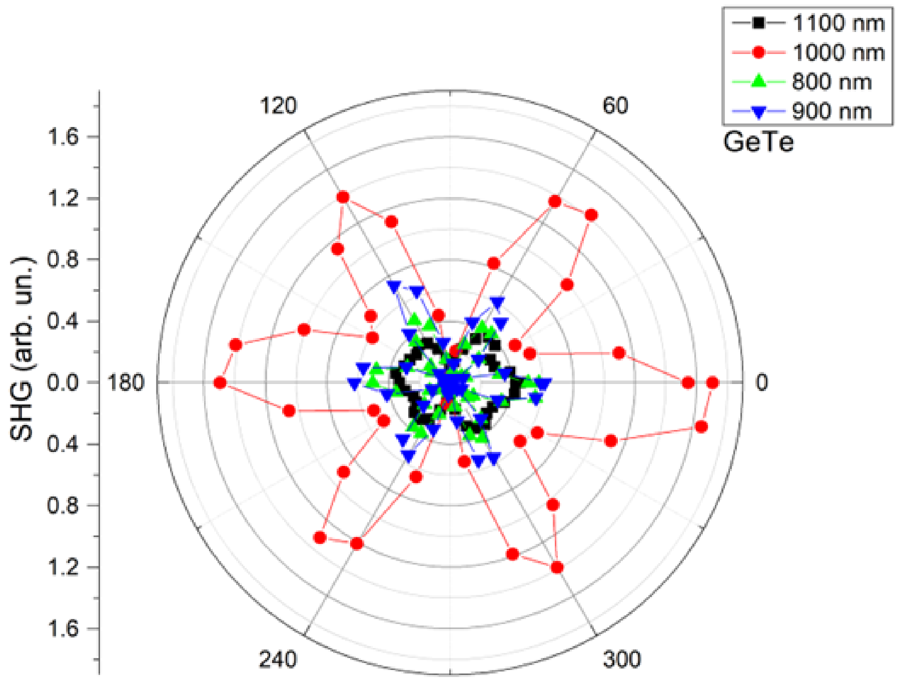


- Ti:Sa :: Titanium Sapphire
- OPA :: Optical parametric resonance
- GT :: Glan Taylor
- WP :: WavePlate
- L1 and L2 :: Lenses
- TF :: Thin Film
- LPF :: Low Pass Filter
- BPF :: Band Pass filter

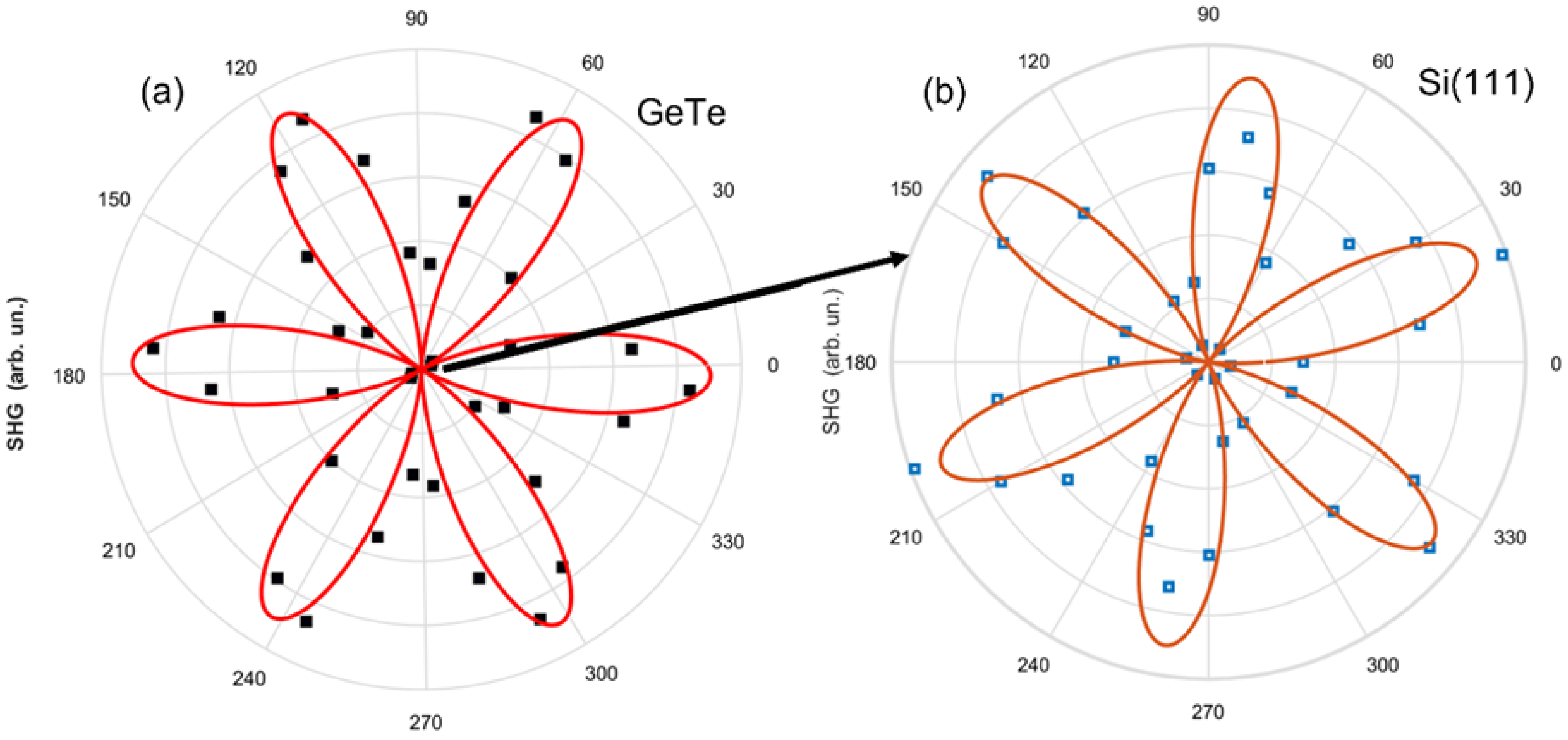
Results


Sample
E(ω)
fs laser
θ
SHG
E(2ω)
x
z
y


GeTe_1000nm
(a)
SHG Intensity (arb. un)
Angle of Incidence (degree)
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0
10
20
30
40
50

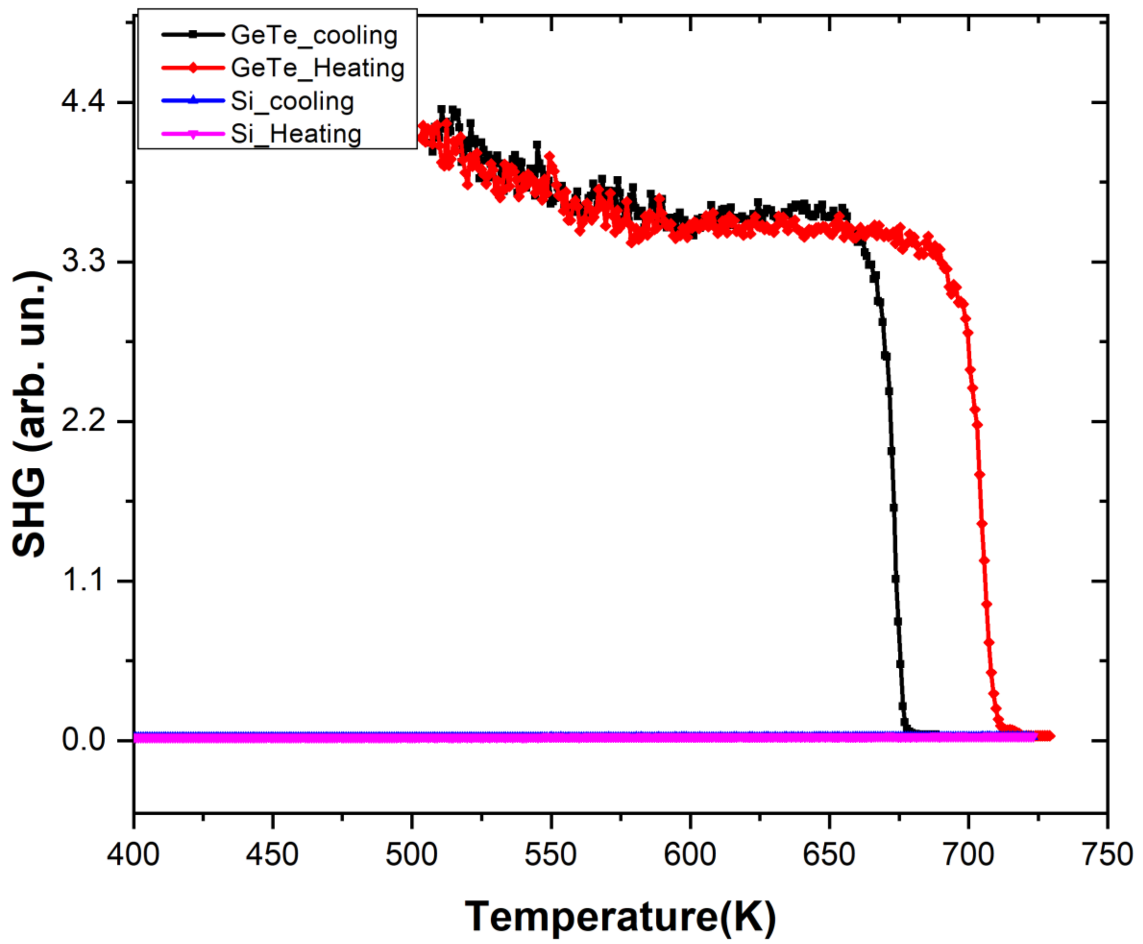

1100 nm
1000 nm
800 nm
900 nm
GeTe
SHG (arb. un.)
120
60
180
0
240
300
1.6
1.2
0.8
0.4
0.0
0.4
0.8
1.2
1.6

SHG Results

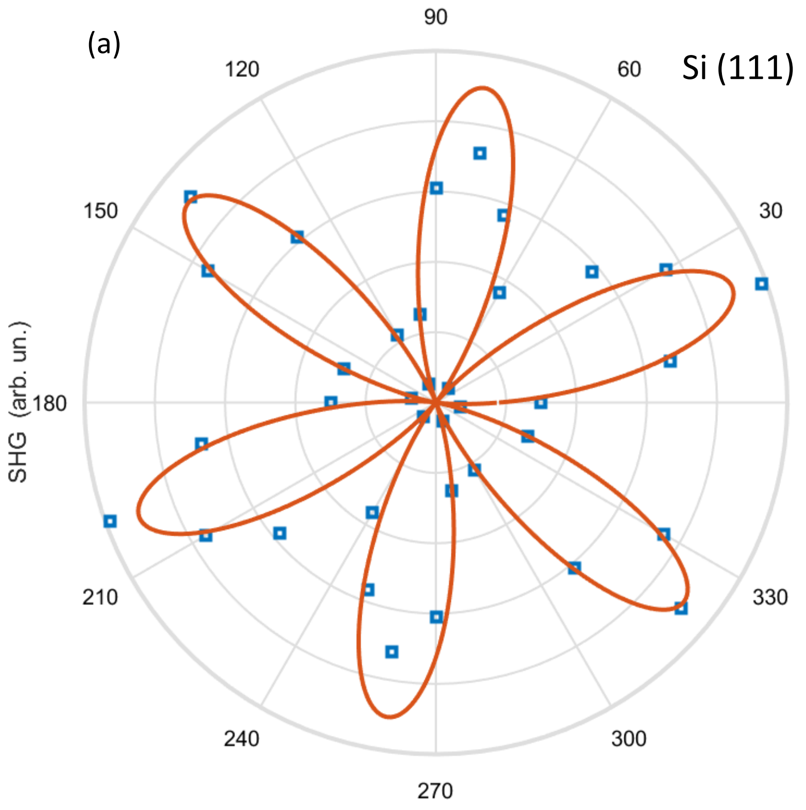


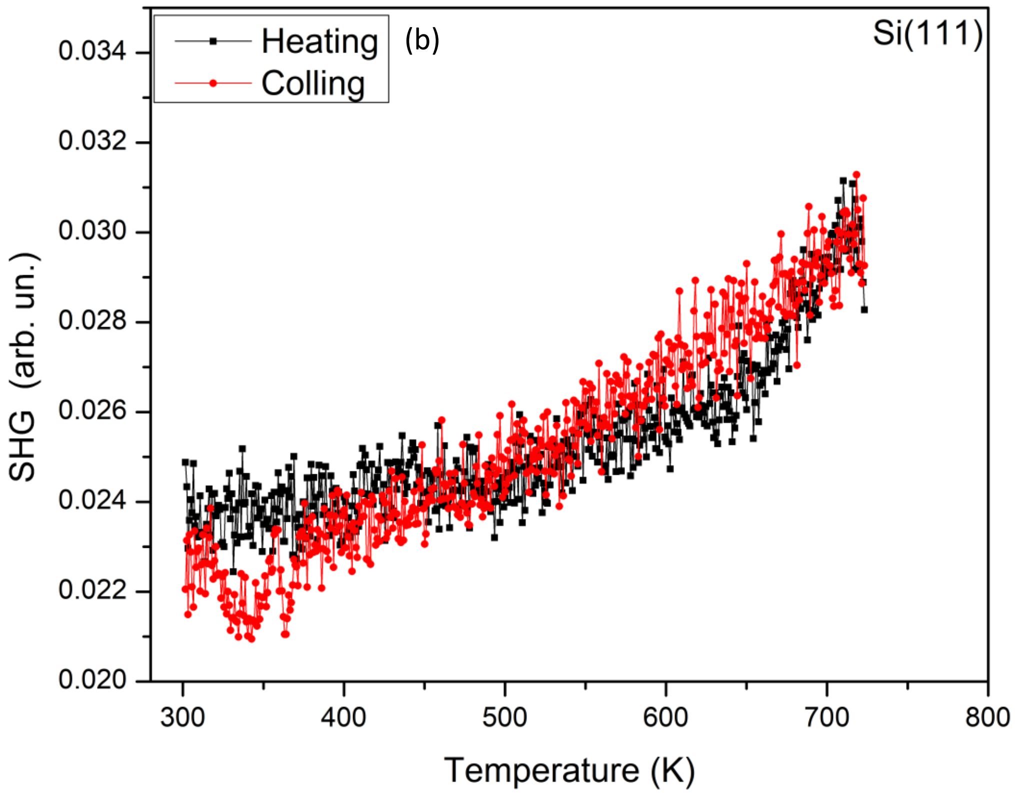
(a)
GeTe
SHG (arb. un.)
90
60
30
0
330
300
270
240
210
180
150
120
(b)
Si(111)
SHG (arb. un.)
90
60
30
0
330
300
270
240
210
180
150
120

Temperature scans of pure GeTe and Silicon



Results

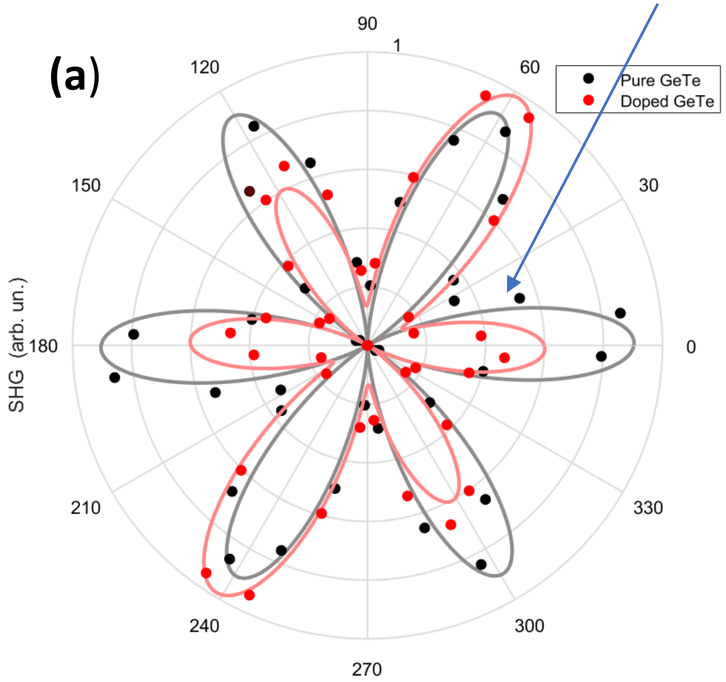

(a)
Si (111)
90
60
30
0
330
300
270
240
210
180
150
120
SHG (arb. un.)

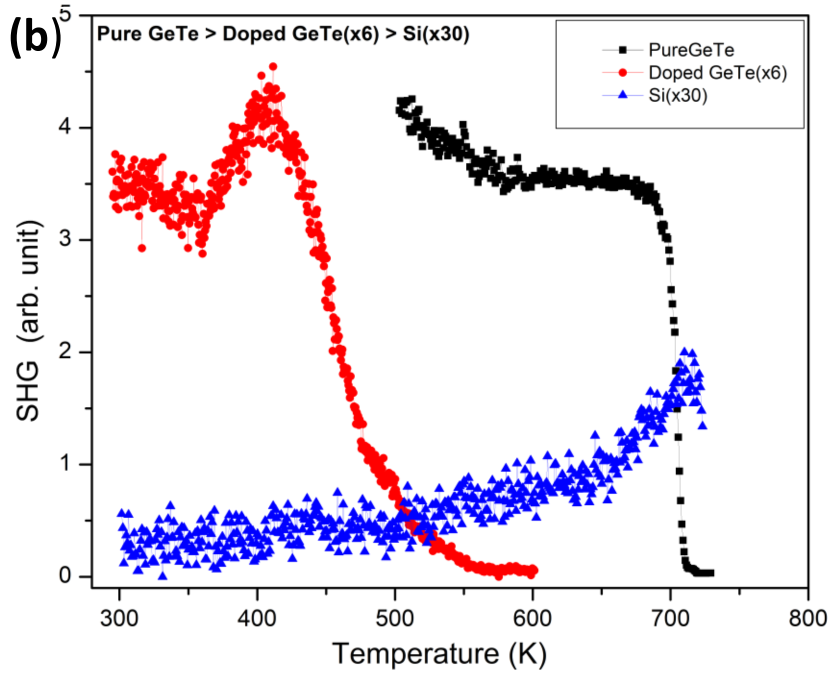

Heating
Colling
(b)
Si(111)
SHG (arb. un.)
0.034
0.032
0.030
0.028
0.026
0.024
0.022
0.020
300
400
500
600
700
800
Temperature (K)

Results

Fit Function

$$I(2\omega = 00{,}900) \propto |A(xxx)\cos 3\phi|^2$$



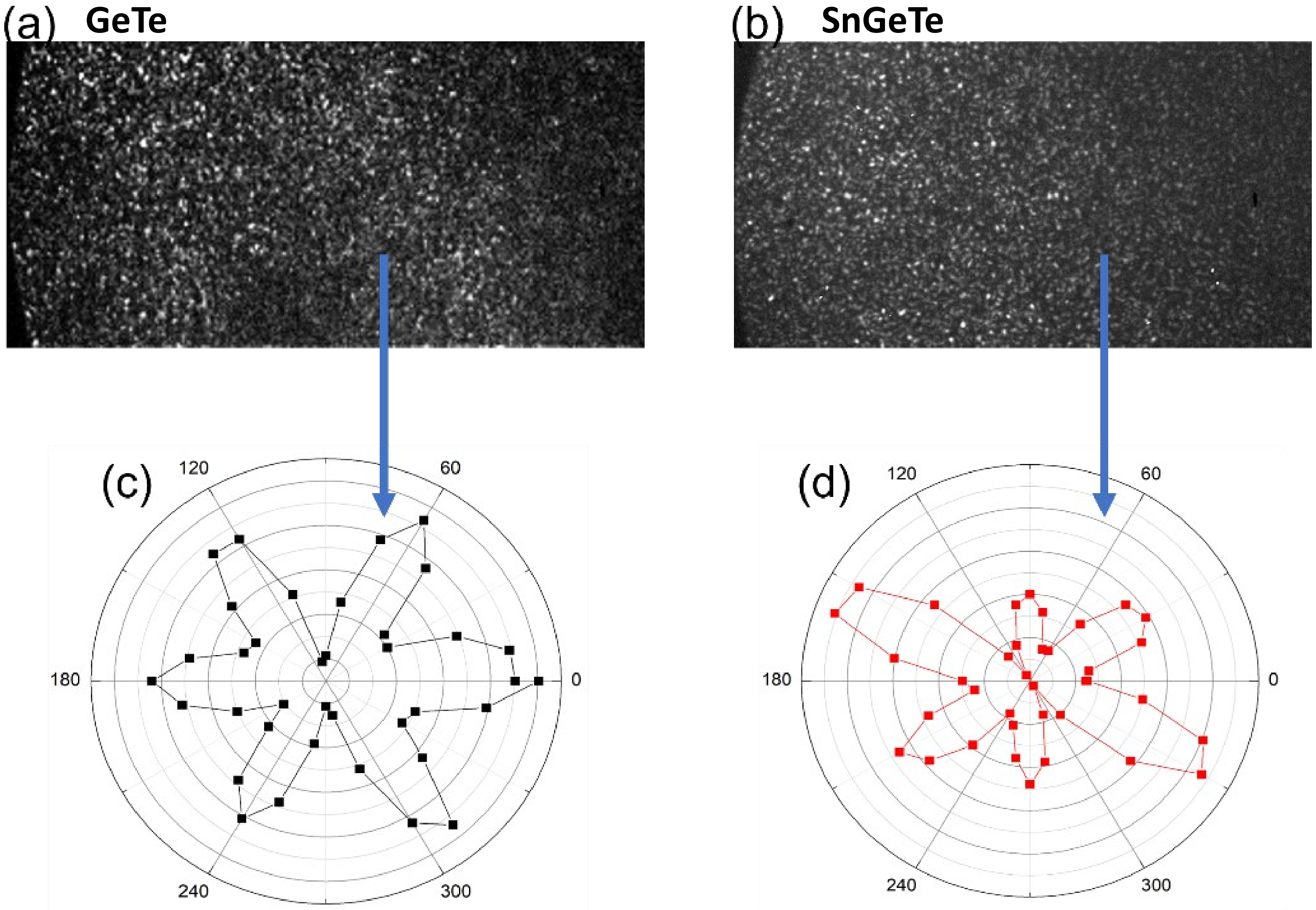


Note : For doped GeTe, the fitting function we used is

$$I(2\omega) \propto 1/4\,A\,(\cos 3\phi) + B\cos(2\beta + \phi))^2$$

Lukas Mendel, NATURE COMMUNICATIONS | (2018) 9:516.

SHG Imaging



Conclusion I

- It is confirmed by the SHG that the LSMO coverage has a significant impact on the HZO FE
- PP and SP are the only components which seems to be affected by the FE in HZO, but the mechanism behind it remains to be clarified
- The SHG cannot be solely generated by LSMO but there is an interfacial contribution from HZO/LSMO interface
- We didn't find any symmetry which can fit our data in electric dipole SHG consideration

Prospective

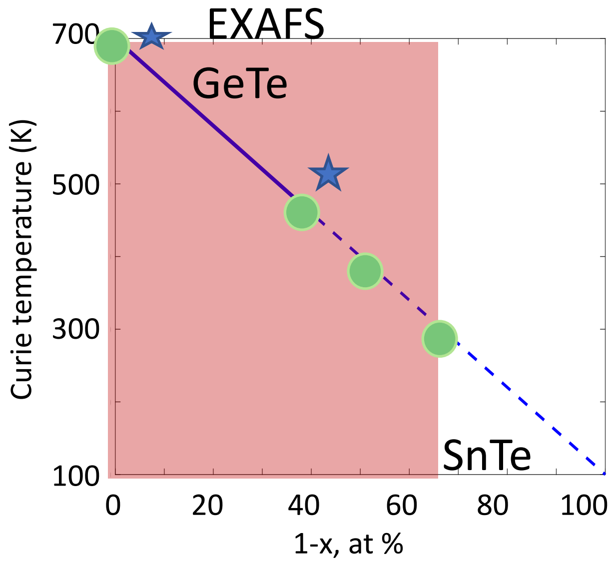
- SHG spectroscopy of HZO samples at higher frequency ?
- SHG Imaging.
- Magnetic measurements.
- High temperature scans.

Conclusion II

- We measure the anisotropies at different wavelengths in order to find the maximum signal
- Temperature dependence measurement shows a higher values of phase transition(T_c = 710 K)for undoped films and 505 K for doped films
- SHG imaging on both pure and doped films shows tiny domains (probably nano-domains)
- The Curie temperature increases with the content of Germanium

Prospects

- *It is also possible to measure SHG image with respect to temperature to see the dependence of domains with time*
- *gating of the ferroelectric semiconductor (switching of the polarization on macroscopic area)*



Adapted from A. Lebedev *et al.*, Ferroelectrics, 298, 189-197 (2004)

Acknowledgements

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Physics of Structured Light And Matter, and of their interaction

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FERROIC

ICMAB Barcelona. Spain
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- Dr. Ignasi Fina

LABORATORY OF MULTIFUNCTIONAL THIN FILMS AND COMPLEX STRUCTURES
INSTITUT DE CIÈNCIA DE MATERIALS DE BARCELONA, ICMAB-CSIC

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