Direct observation of controlled strain-induced second harmonic generation in a $Co_{0.25}Pd_{0.75}$ thin film on a $Pb(ZrTi)O_3$ substrate

Appl. Phys. Lett. ***90****, 044108 (2007), J. Jeong etc.*

The authors have observed strain-induced second harmonic generation (SHG) signals from a $Co_{0.25}Pd_{0.75}$ alloy film deposited on a lead zirconate titanate (PZT) substrate. The strain in the sample was controlled by the inverse piezoelectric effect. The authors demonstrate that it is possible to separate the strain contribution to the SHG signal from the crystallographic contribution and that from the electric polarization in PZT. An estimate of the value of the nonlinear photoelastic tensor components is in very good agreement with previous calculations.

발표자 : 전윤남

24th July, 2008

Soft-Matter Optical Spectroscopy

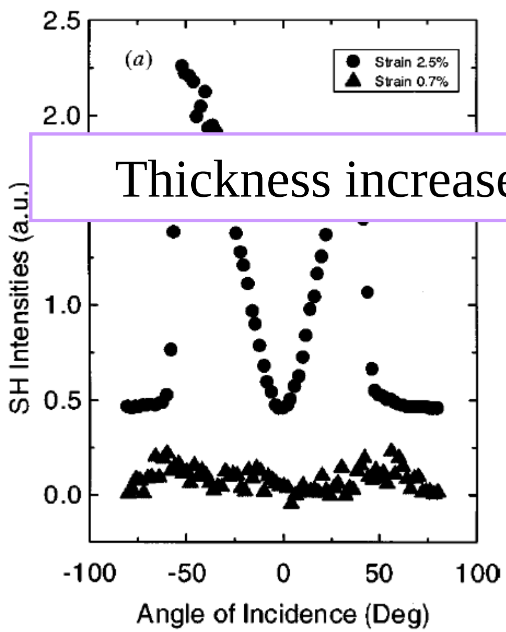
Introduction : strain induced SHG

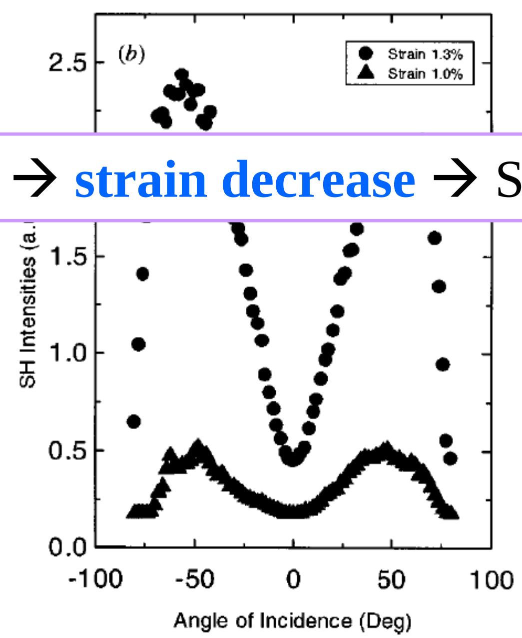
U. C. Oh et al., Appl. Phys. Lett., **76**, 1461 (2000)

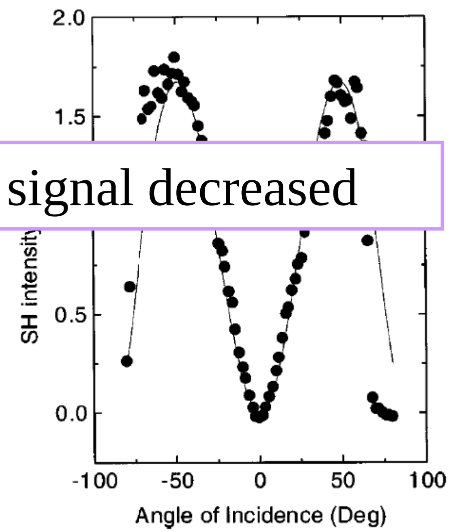
Epitaxial grown $Ba_{0.48}Sr_{0.52}TiO_3$ thin films (300 Å & 1650 Å) on the (001) MgO

→ **<u>centrosymmetric</u>** cubic (m3m crystal symmetry)

SHG should be absent







Thickness increase → **strain decrease** → SHG signal decreased

Maker fringe (1064 nm Nd:YAG laser)

tetragonal distortion

(**4mm** symmetry)

Introduction : strain induced SHG

The symmetry of the twin boundaries of ZnO epitaxial film was detected with reflective second harmonic generation (RSHG). The twin boundaries exhibit mirror symmetry with a polar configuration across the boundary plane and yield a nonvanishing polar contribution to RSHG. The nonvanishing second-order susceptibility supports the notion that the measured RSHG originates from the planar defect, which depends on the residual stress in the thin film. We analyzed our RSHG result by correlating the macroscopic data from optic probe with the microscopic data from tunneling electron microscope. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2891334]

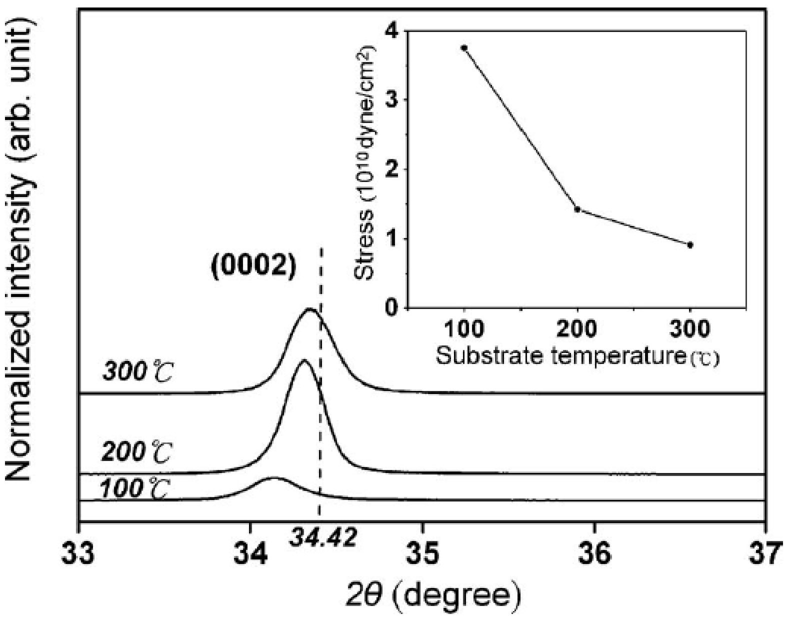
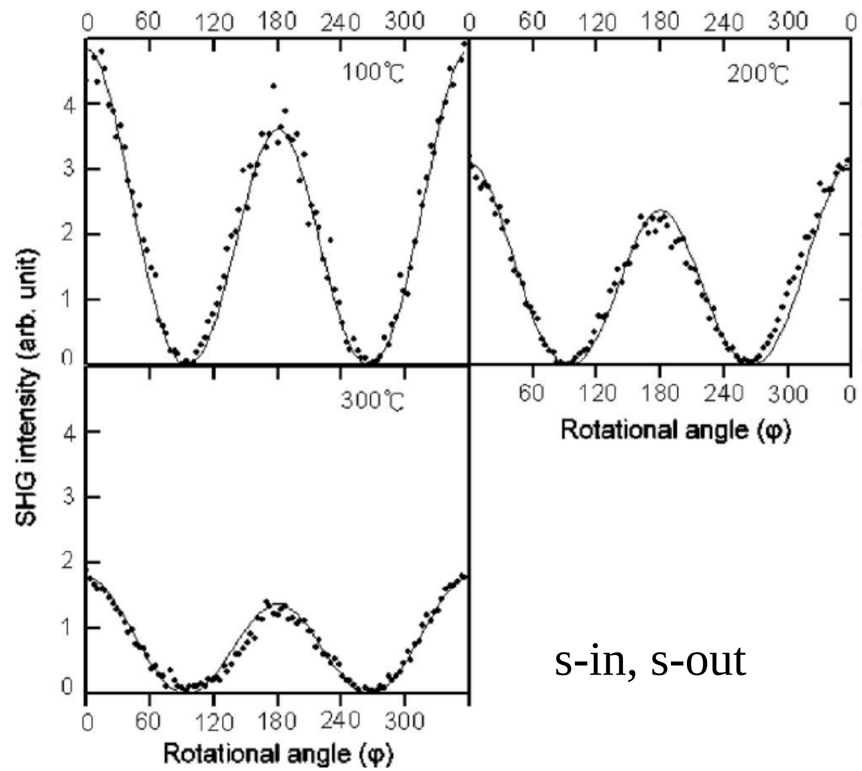


FIG. 1. The x-ray patterns of ZnO films deposited on Si(111) at different substrate temperature. The inset shows the stress calculated by the XRD profiles and peak shifts of the ZnO (0002) peak.



s-in, s-out

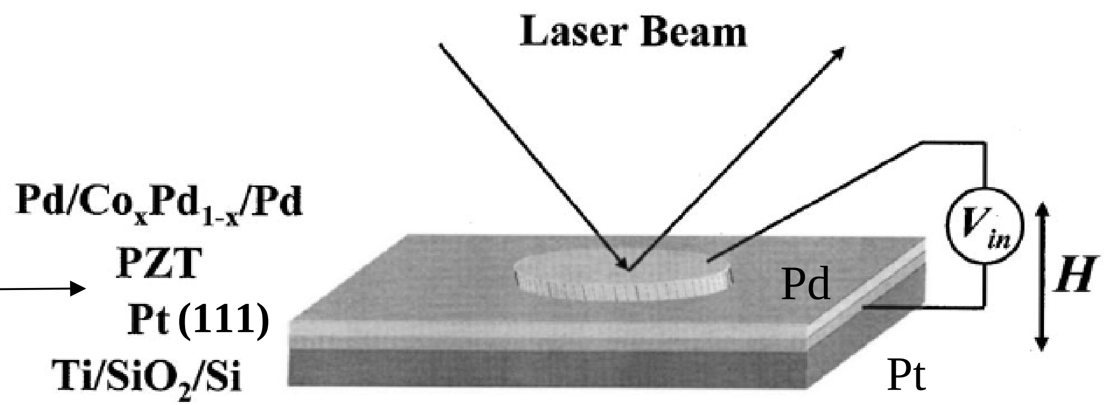
K. Lo et al., Appl. Phys. Lett., **92**, 091909 (2008)

Sample preparation

J. Lee et al., Appl. Phys. Lett., **82**, 2458 (2003)

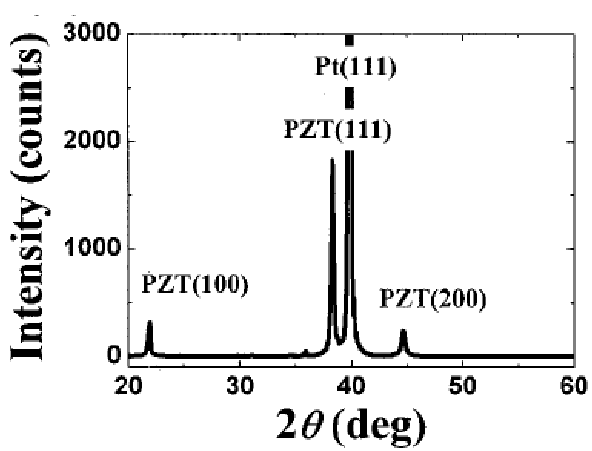
deposition

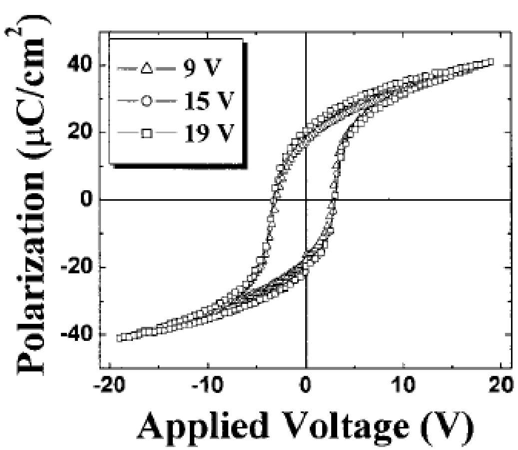
multiple spin coating
(annealing : 650 °C)



PZT ~ 1 μm (~120-200 nm * 6 times)

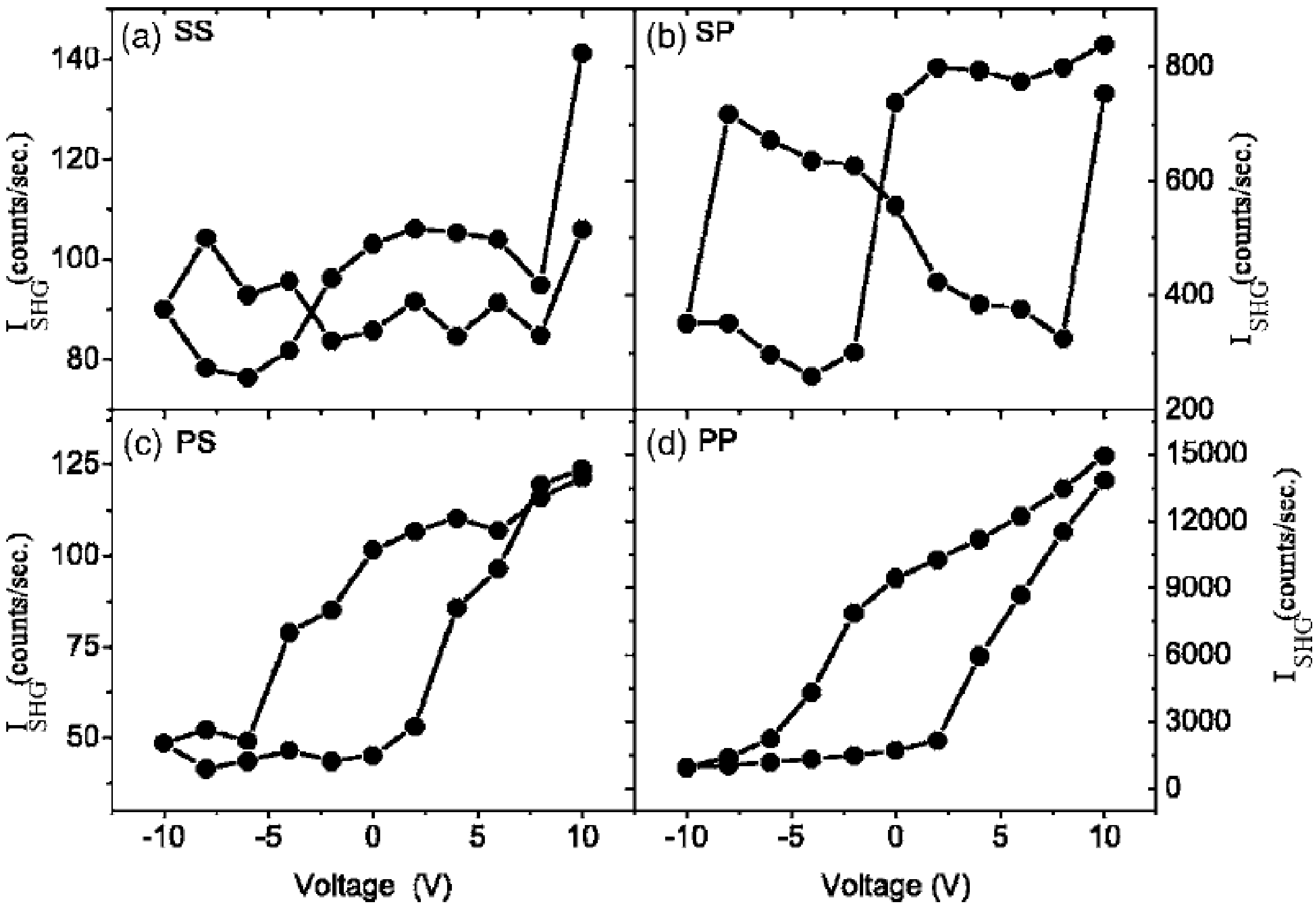
Pd (30 Å)/ Co$_x$Pd$_{1-x}$ (30 Å)/ Pd (30 Å) (buffer layer)





Soft-Matter Optical Spectroscopy

Strain induced SHG data



Soft-Matter Optical Spectroscopy

Theory

$$\boldsymbol{I(2\omega)} \propto |P^{NL}(2\omega)|^2 \propto |\boldsymbol{\chi^{(2)}_{ijk}}|^2 \boldsymbol{I^2(\omega)}$$

with $P_i^{NL}(2\omega) = \chi^{(2)}_{ijk}(-2\omega{:}\ \omega,\ \omega)\, E_j(\omega)\, E_k(\omega)$

SHG : (1) **crystallographic** term (depend on the symmetry)

(2) **electric polarization** ρ of the ferroelectric substrate

(3) **strain induced** term

$$\chi^{(\mathrm{eff})}_{ijk} = \boldsymbol{\chi^{(2,0)}_{ijk}} + \boldsymbol{p_{ijklm} u_{lm}} + \boldsymbol{\chi^{(3,0)}_{ijkl} P_l}$$

$\chi^{(2,0)}_{ijk}$: purely electronic NOS (nonlinear optical susceptibility) tensor

$\chi^{(3,0)}_{ijkl}$: third-order NOS tensor

Soft-Matter Optical Spectroscopy

Theory : strain induced term

$$\chi^{(\mathrm{eff})}_{ijk} = \chi^{(2,0)}_{ijk} + p_{ijklm} u_{lm} + \chi^{(3,0)}_{ijkl} P_l$$

$$P_i = P_i^{(0)} + k_{ij} E_j^{int}$$

k_{ij} : dielectric susceptibility

E^{int} : internal electric field (U/t_f)

U : applied external voltage,

t_f : total thickness of the film

$P_i^{(0)}$: spontaneous polarization of the ferroelectric substrate

$\boldsymbol{p_{ijklm}}$: NPE (nonlinear photoelastic) tensor

$u_{lm} = d_{lmn} E_n^{int}$ (**strain tensor**)

(d_{lmn} : tensor describing the inverse piezoelectric effect)

Theory : Photoelastic tensor

Crystal symmetry	Nonzero components of p_{ijklm}
$4mm$	$yxxyz=xyyxz,\ yxyxz=xxyyz,\ yxzxy=xyzyx,\ yyyyz=xxxxz,\ yyzxx=xxzyy$ $yyzyy=xxzxx,\ yyzzz=xxzzz,\ yzzyz=xzzxz,\ zxyyx,\ zyyxx=zxxyy$ $zyyyy=zxxxx,\ zyyzz=zxxzz,\ zyzyz=zxzxz,\ zzzyy=zzzxx,\ zzzzz$
$3m$	$xxxyy,\ xxxx,\ yyyyx=yxxxy=-\frac{1}{2}xxxxx+\frac{1}{2}xxxyy$ $xyyy=xyyxx=xxxx,\ yyxyy=yyxxx=-\frac{1}{2}xxxxx-\frac{1}{2}xxxyy$ $xyxyz=-yyxxz,\ xxxxz=-yyyyz,\ yxxyz=xyyxz=xxxxz-2xyxz$ $xxxzz=-yyxzz=-xyyzz,\ xyzxy=yxzyx=\frac{1}{2}xxzxx-\frac{1}{2}xxzyy$ $xxzxx=yyzyy,\ yyzzx=yxzzy=xyzzy=-xxzxz,\ xzzxx=-yzzyx=-xzzyy$ $zxyxy=\frac{1}{2}zxxxx-\frac{1}{2}zxxyy,\ zxxyy=zyyxx,\ zzzyy=zzzxx$ $zxxxz=-zyyxz=-zxyyz,\ zxzxx=-zyzyx=-zxzyy,\ yyzzz=xxzzz$ $xxzyy=yyzxx,\ yzzyz=xzzxz,\ zyyzz=zxxzz,\ zyzyz=zxzxz,\ zzzzz$

J. Jeong et al., Phys. Rev. B, **62**, 13455 (2000)

Theory : Point group

3m (C_{3v}) point group :

xzx = yzy,
xxz = yyz,
zxx = zyy,
zzz,
yyy = -yxx = -xxy = -xyx

$$d_{ij} = \begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{15} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix}$$

C. Jung et al., Appl. Optics, **39**, 5142 (2000)

ex) $LiNbO_3$: d_{22} = 7.4, d_{31} = 14, d_{33} = 98

R. W. Boyd, *Nonlinear Optics*, Academic press (1992)

Components of strain tensor, u_{lm}	Components of second-order NOS tensor, $\chi^{(2)}_{ijk}$: 4mm	Components of second-order NOS tensor, $\chi^{(2)}_{ijk}$: 3m
xx,yy,zz	*xxz*=*xzx*, *yyz*=*yzy*, *zxx*, *zyy*, *zzz*,	*xxx*, *xxz*=*xzx*, *xyy*, *yxy*=*yyx*, *yyz*=*yzy*,
xz=*zx*	**xxx**, **xyy**, **xzz**, **yxy**=**yyx**, **zxz**=**zzx**	*zxx*, **xzz**, **zxz**=**zzx**, *zyy*, **zzx**=**zxz**, *zzz*
yz=*zy*	**xxy**=**xyx**, **yxx**, **yyy**, **yzz**, **zyz**=**zzy**	**xxy**=**xyx**, **xyz**=**xzy**, **yxx**, **yxz**=**yzx**,
xy=*yx*	**xyz**=**xzy**, **yxz**=**yzx**, **zxy**=**zyx**	**zxy**=**zyx**, **zyz**=**zzy**, **yyy**, **yzz**

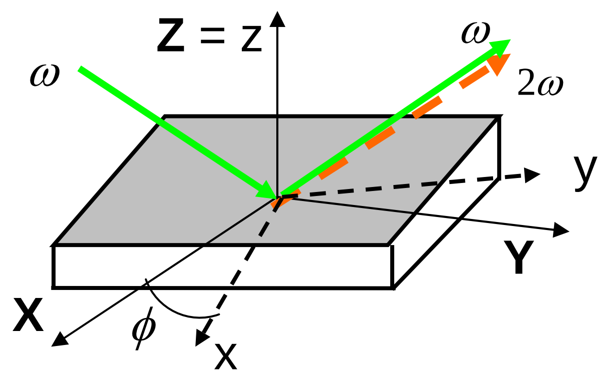
J. Jeong et al., Phys. Rev. B, **62**, 13455 (2000)

Theory : Effective susceptibility

Laboratory coordinate (X, Y, Z) :

azimuthal angle (ϕ)

Molecular coordinate (x, y, z) :



$$\chi_{\text{eff}}^{(2)} = [\hat{\mathbf{e}}(\omega)\cdot\mathbf{L}(\omega)]\cdot\chi^{(2)}:[\mathbf{L}(\omega_1)\cdot\hat{\mathbf{e}}(\omega_1)][\mathbf{L}(\omega_2)\cdot\hat{\mathbf{e}}(\omega_2)]$$

ω

$$\begin{cases} \vec{L}\cdot\vec{S} = (L_{yy}'\sin\phi,\ L_{yy}'\cos\phi,\ 0) \\ \vec{L}\cdot\vec{P} = (L_{xx}'\cos\theta\cos\phi,\ -L_{xx}'\cos\theta\sin\phi,\ L_{zz}'\sin\theta) \end{cases}$$

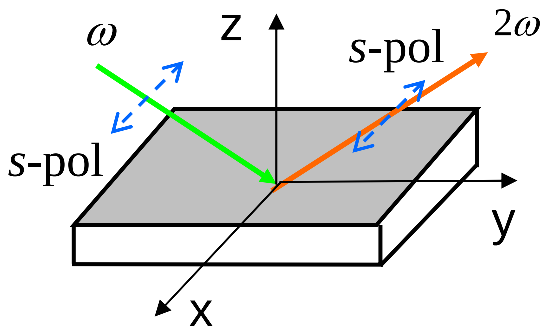
2ω

$$\begin{cases} \vec{L}\cdot\vec{S} = (L_{yy}\sin\phi,\ L_{yy}\cos\phi,\ 0) \\ \vec{L}\cdot\vec{P} = (-L_{xx}\cos\theta\cos\phi,\ L_{xx}\cos\theta\sin\phi,\ L_{zz}\sin\theta) \end{cases}$$

X. Zhuang et al., Phys. Rev. B, **59**, 12632 (1999)

Theory : Effective susceptibility

$$\chi_{\text{eff}}^{(2)} = [\hat{\mathbf{e}}(\omega)\cdot\mathbf{L}(\omega)]\cdot\chi^{(2)}:[\mathbf{L}(\omega_1)\cdot\hat{\mathbf{e}}(\omega_1)][\mathbf{L}(\omega_2)\cdot\hat{\mathbf{e}}(\omega_2)]$$



ω

$$\begin{cases} \vec{L}\cdot\vec{S} = (L_{yy}'\sin\phi, L_{yy}'\cos\phi, 0) \\ \vec{L}\cdot\vec{P} = (L_{xx}'\cos\theta\cos\phi, -L_{xx}'\cos\theta\sin\phi, L_{zz}'\sin\theta) \end{cases}$$

2ω

$$\begin{cases} \vec{L}\cdot\vec{S} = (L_{yy}\sin\phi, L_{yy}\cos\phi, 0) \\ \vec{L}\cdot\vec{P} = (-L_{xx}\cos\theta\cos\phi, L_{xx}\cos\theta\sin\phi, L_{zz}\sin\theta) \end{cases}$$

$$\chi_{sss}^{(2)} = \chi_{yyy}^{(2)} L_{yy}\cos\phi\, L_{yy}^2{}'\cos^2\phi + \chi_{yxx}^{(2)} L_{yy}\cos\phi\, L_{yy}^2{}'\sin^2\phi + \chi_{xxy}^{(2)} L_{yy}\sin\phi\, L_{yy}^2{}'\sin\phi\cos\phi + \chi_{xyx}^{(2)} L_{yy}\sin\phi\, L_{yy}^2{}'\sin\phi\cos\phi$$

$$\boxed{\chi_{sss}^{(2)} = \chi_{yyy}^{(2)} L_{yy} L_{yy}^2{}'\cos\phi\,(\cos^2\phi - 3\sin^2\phi)}$$

xzx = yzy,
xxz = yyz,
zxx = zyy,
zzz,

yyy = -yxx = -xxy = -xyx

X. Zhuang et al., Phys. Rev. B, **59**, 12632 (1999)

Soft-Matter Optical Spectroscopy

Theory : Effective susceptibility

$$\chi^{(2)}_{\text{eff}} = [\hat{\mathbf{e}}(\omega)\cdot\mathbf{L}(\omega)]\cdot\chi^{(2)}:[\mathbf{L}(\omega_1)\cdot\hat{\mathbf{e}}(\omega_1)][\mathbf{L}(\omega_2)\cdot\hat{\mathbf{e}}(\omega_2)]$$

ω
$$\begin{cases} \vec{L}\cdot\vec{S} = (L_{yy}'\sin\phi,\ L_{yy}'\cos\phi,\ 0) \\ \vec{L}\cdot\vec{P} = (L_{xx}'\cos\theta\cos\phi,\ -L_{xx}'\cos\theta\sin\phi,\ L_{zz}'\sin\theta) \end{cases}$$

2ω
$$\begin{cases} \vec{L}\cdot\vec{S} = (L_{yy}\sin\phi,\ L_{yy}\cos\phi,\ 0) \\ \vec{L}\cdot\vec{P} = (-L_{xx}\cos\theta\cos\phi,\ L_{xx}\cos\theta\sin\phi,\ L_{zz}\sin\theta) \end{cases}$$

xzx = yzy,
xxz = yyz,
zxx = zyy,
zzz,
yyy = -yxx = -xxy = -xyx

$$\chi^{(2)}_{spp} = \chi^{(2)}_{yyy}\ L_{yy}L^2_{xx}'\cos\phi\sin^2\theta(3\sin^2\phi-\cos^2\phi)$$

$$\chi^{(\text{eff})}{}_{ijk} = \chi^{(2,0)}{}_{ijk} + p_{ijklm}u_{lm} + \chi^{(3,0)}{}_{ijkl}\,P_l$$

$$\begin{cases} \chi^{(eff)}_{sss} = \chi^{(2,0)}_{yyy}\ L_{yy}L^2_{yy}'\cos\phi(\cos^2\phi-3\sin^2\phi) \\ \chi^{(eff)}_{spp} = \chi^{(2,0)}_{yyy}\ L_{yy}L^2_{xx}'\cos\phi\sin^2\theta(3\sin^2\phi-\cos^2\phi) \end{cases}$$

Components of strain tensor, u_{lm}
xx, yy, zz
$xz = zx$
$yz = zy$
$xy = yx$

Theory : Effective susceptibility

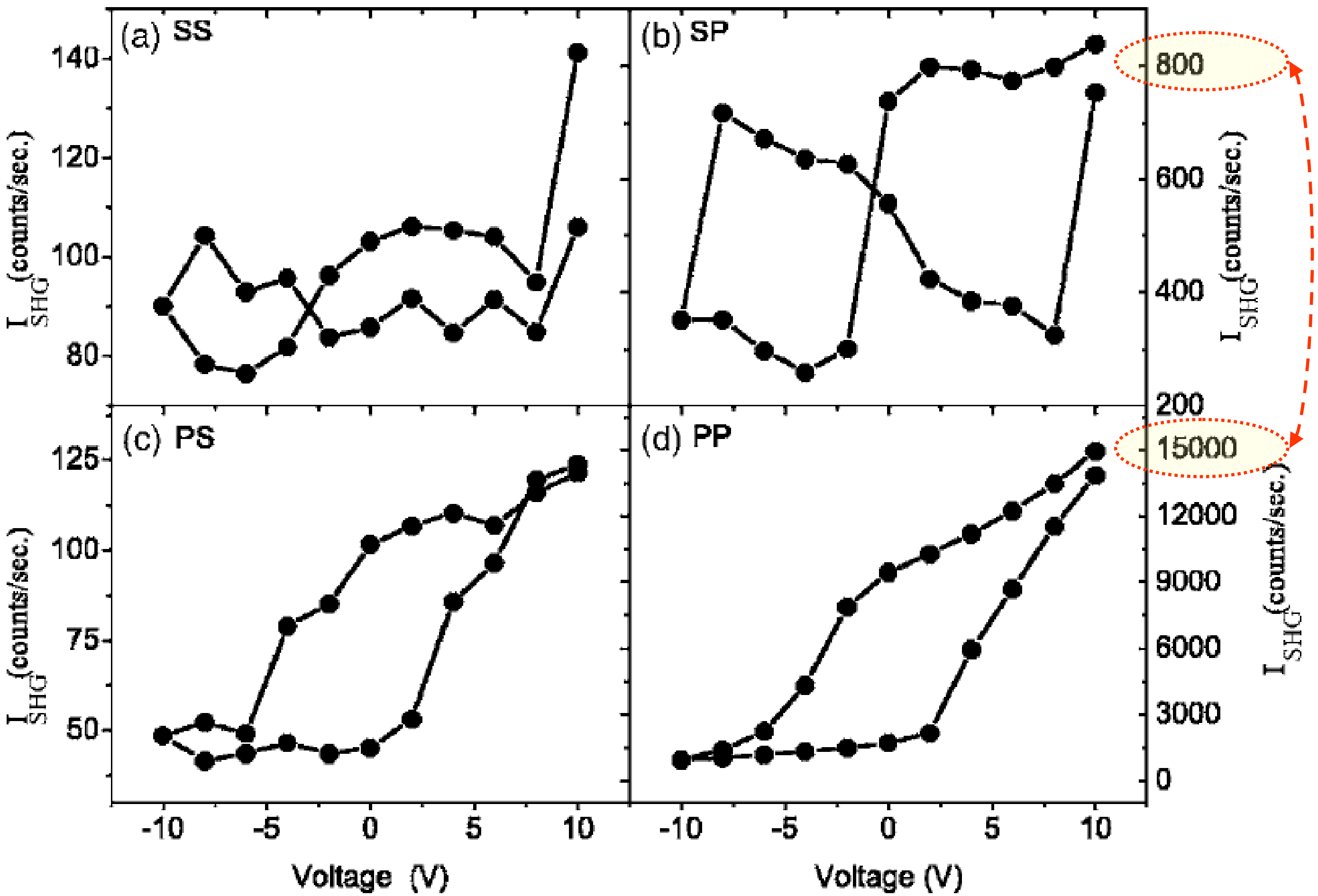
$$\chi^{(\text{eff})}_{s;ss} = \chi^{(2,0)}_{yyy},$$

$$\chi^{(\text{eff})}_{p;ss} = \{(\chi^{(3,0)}_{xyyz}\mathcal{P}_z + p_{xyyzz}u_{zz})^2 + (\chi^{(2,0)}_{zyy} + \chi^{(3,0)}_{zyyz}\mathcal{P}_z + p_{zyyxx}u_{xx} + p_{zyyyy}u_{yy} + p_{zyyzz}u_{zz})^2\}^{1/2},$$

$$\chi^{(\text{eff})}_{s;pp} = \chi^{(2,0)}_{yxx}\cos^2\theta,$$

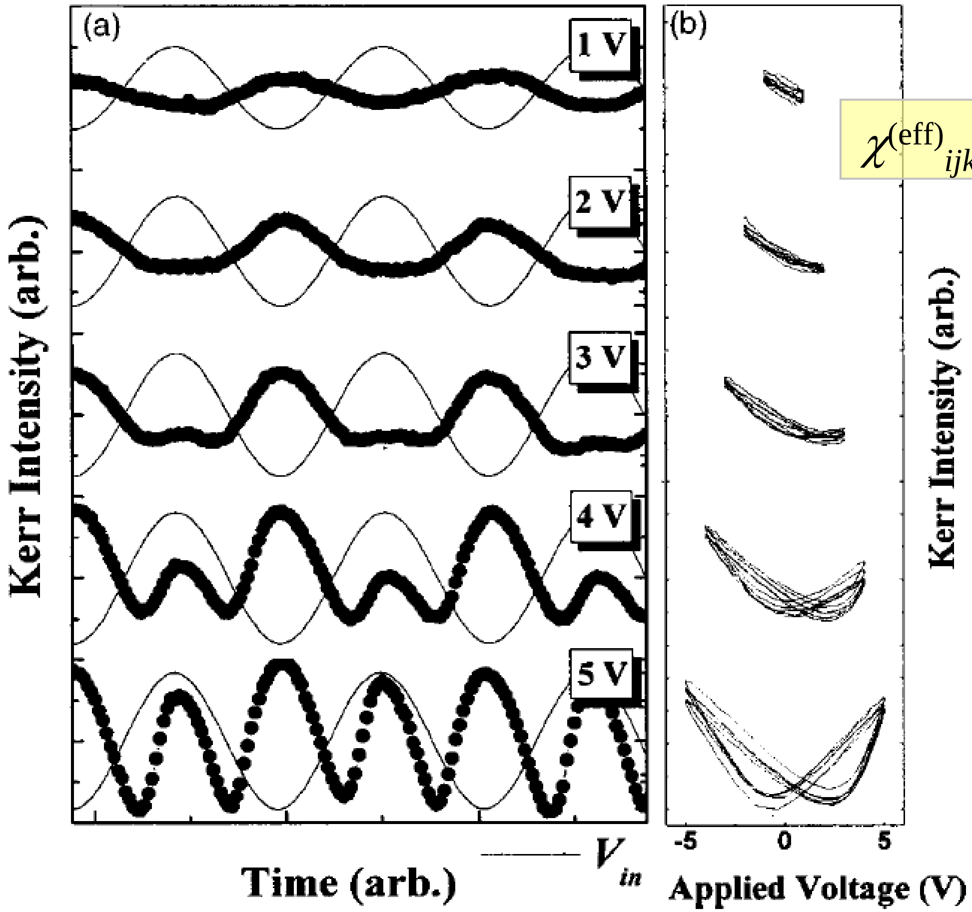
$$\chi^{(\text{eff})}_{p;pp} = \{[(\chi^{(3,0)}_{xxxz}\mathcal{P}_z + p_{xxxxx}u_{xx} + p_{xxxyy}u_{yy} + p_{xxxzz}u_{zz})\cos^2\theta + (\chi^{(2,0)}_{xzx} + \chi^{(2,0)}_{xxz} + \chi^{(3,0)}_{xxzz}\mathcal{P}_z + 2p_{xzxxx}u_{xx} + 2p_{xzxyy}u_{yy} + 2p_{xzxzz}u_{zz})\cos\theta\sin\theta + (p_{xzzxx}u_{xx} + p_{xzzyy}u_{yy})\sin^2\theta]^2 + [(\chi^{(2,0)}_{zxx} + \chi^{(3,0)}_{zxxz}\mathcal{P}_z + p_{zxxxx}u_{xx} + p_{zxxyy}u_{yy} + p_{zxxzz}u_{zz})\cos^2\theta + (\chi^{(2,0)}_{zzz} + p_{zzzzz}u_{zz})\sin^2\theta]^2\}^{1/2},$$

Strain induced SHG data



$\chi^{(2,0)}_{zzz}$ is dominant

Contribution to the SHG signal



$$\chi^{(eff)}_{ijk} = \chi^{(2,0)}_{ijk} + p_{ijklm}u_{lm} + \chi^{(3,0)}_{ijkl}\, P_l$$

$U_c = 3$ V → $P = 0$

$$\chi^{(eff)}_{ijk} = \chi^{(2,0)}_{ijk} + p_{ijklm}u_{lm}$$

$$= \chi^{(2,0)}_{ijk} + p_{ijklm}\, d_{lmn}\, E_n^{int}$$

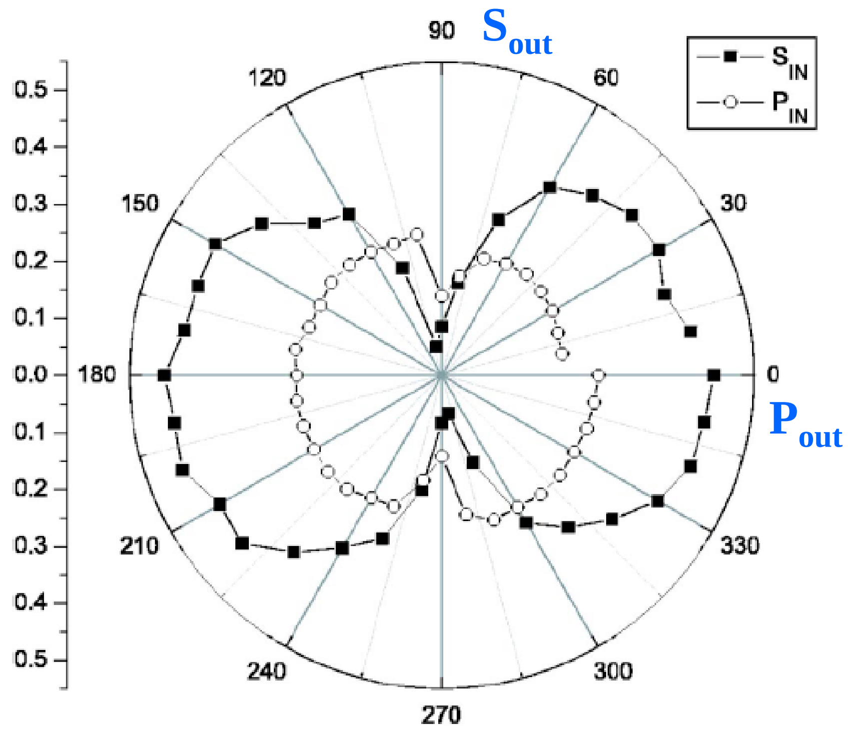
J. Lee et al., Appl. Phys. Lett., **82**, 2458 (2003)

Soft-Matter Optical Spectroscopy

Strain induced SHG

$$\chi^{(\mathrm{eff})}_{ijk} = \chi^{(2,0)}_{ijk} + p_{ijklm}\, d_{lmn}\, E_n^{int}$$

$$\begin{cases} I^{up}(2\omega) : \chi^{(\mathrm{eff})}_{ijk} = \chi^{(2,0)}_{ijk} + p_{ijklm}\, d_{lmn}\, E_n^{int} \\ I^{down}(2\omega) : \chi^{(\mathrm{eff})}_{ijk} = \chi^{(2,0)}_{ijk} - p_{ijklm}\, d_{lmn}\, E_n^{int} \end{cases}$$



$$A_{SHG} = \frac{I^{\uparrow}(2\omega) - I^{\downarrow}(2\omega)}{I^{\uparrow}(2\omega) + I^{\downarrow}(2\omega)}$$

$$\propto \frac{\left| p^{NL,eff}\, d_{zzz} E_z^{\mathrm{int}} \right|}{\left| \chi^{(2,0)} \right|} \cos\varphi$$

(ϕ : phase between the crystallographic and NPE contribution)