Second Harmonic Spectroscopy Study of Silicon Nanocrystals Embedded in SiO_2

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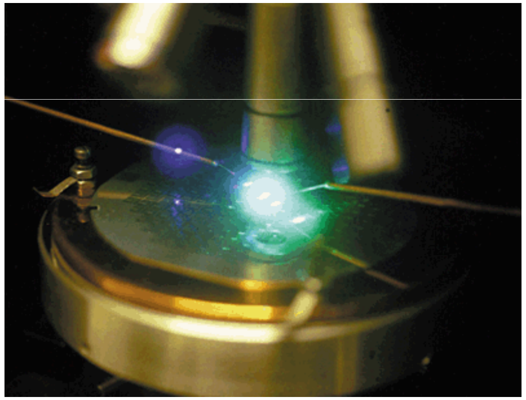
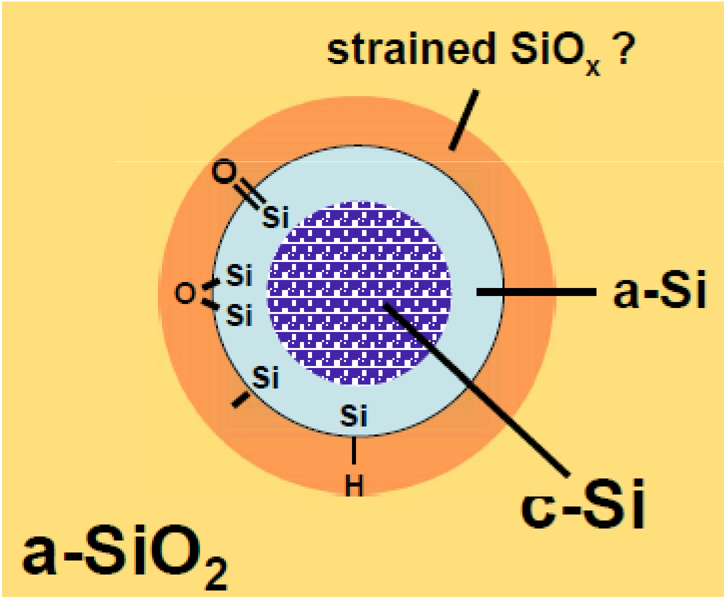
Si nanocrystals have properties & applications different from those of bulk Si

"Si lasers start to take shape"

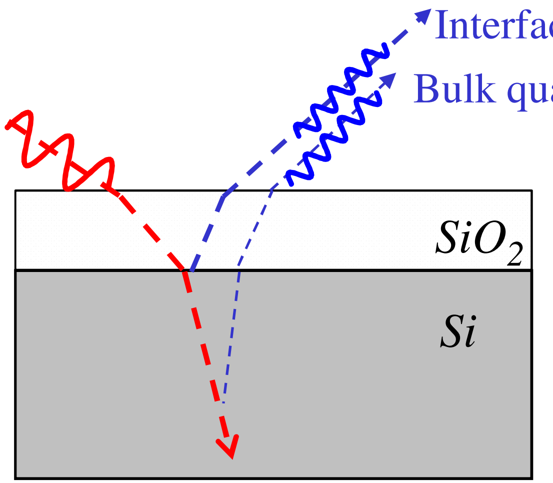


Observation of optical gain in Si nanocrystals embedded in SiO_2
*Pavesi et al., Nature **408**, 440 (2000)*



Those interesting properties originate at Si NC/SiO2 interfaces
SHG has a reputation for being interface-specific

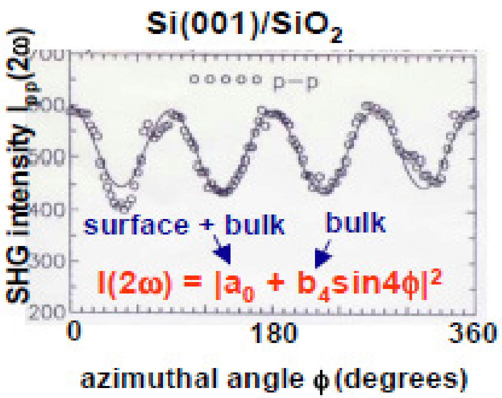
Interface and bulk contributions to SHG from planar surfaces are never sperated with full rigor

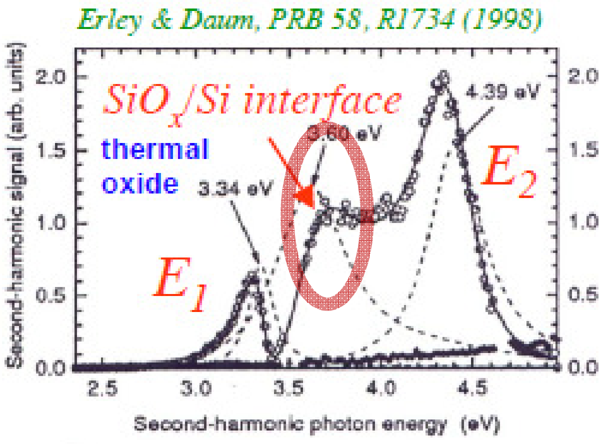


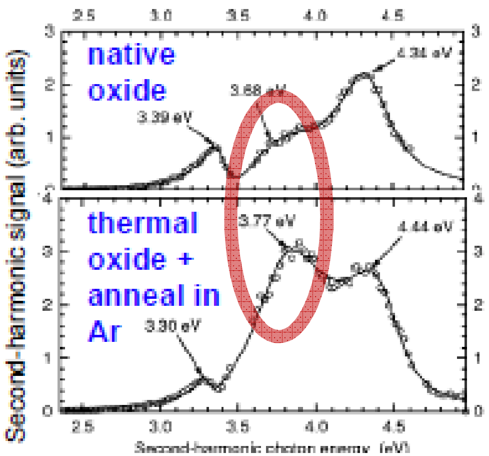
*J.E. Sipe et. al., Phys. Rev. B **35**, 1129 (1987)*

Empirical separation of surface & bulk contribution is usually based on:

1. Azimuthal anisotropy
2. Spectroscopy study
3. Interface modification



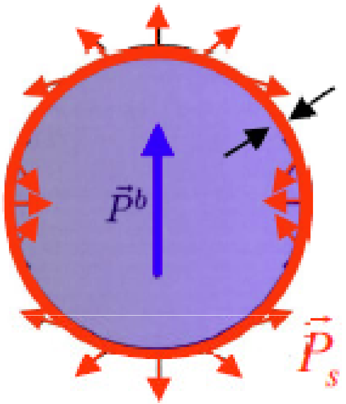




Similar bulk/interface ambiguity in SHG from Si NCs must be distinguished empirically

Mochan et. al., Phys. Rev. B ***68****, 085318 (2003)*

single nanoparticle:



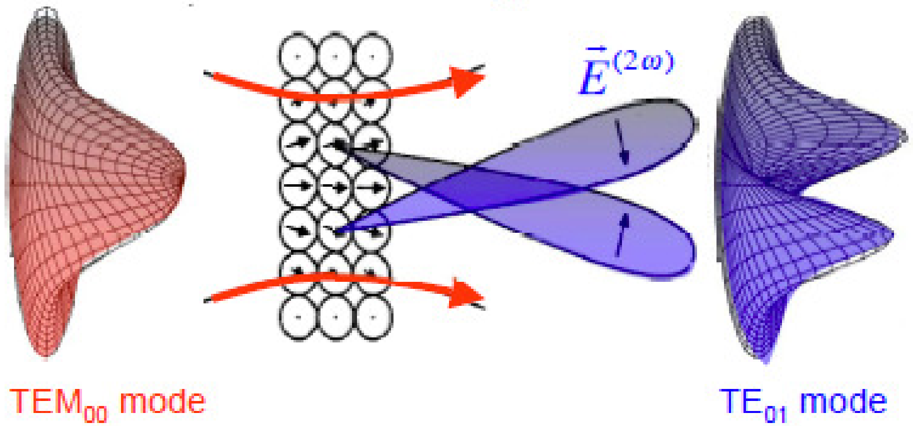
From symmetry alone,

$$\vec{P}^b(\vec{r}) = \gamma\nabla E^2 + \delta' \vec{E}\cdot\nabla\vec{E}$$

$$\vec{P}^s(\vec{r}) = \chi^s_{ijk}(a,b,f)F_jF_k,$$

assuming $l << r_{NC} << \lambda$

uniform nano-composite:



$$\vec{P}^{NL} = \Delta' \vec{E}\cdot\nabla\vec{E}$$

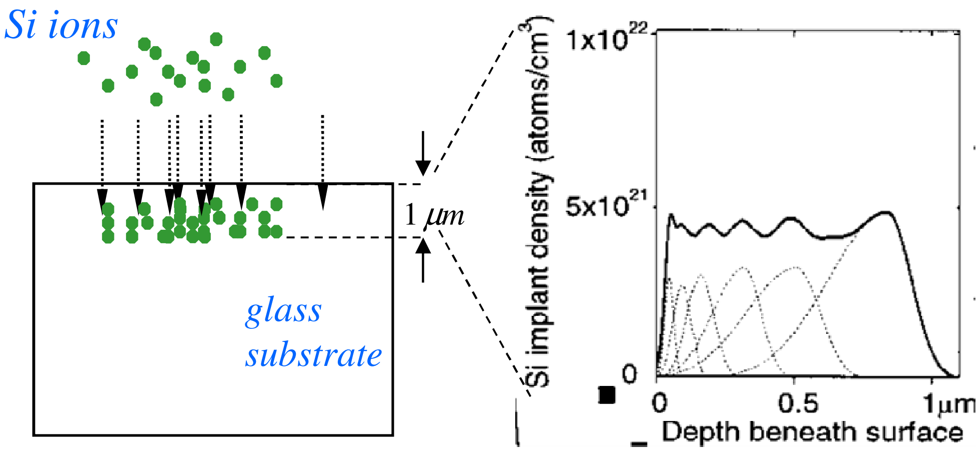
$$\begin{aligned}\Delta' \equiv n_{NC}[\gamma_e(\delta',\gamma,a,b,f) \\ -\gamma_m(\delta',\gamma,a,b,f) \\ -\gamma_q(a,b,f)/6]\end{aligned}$$

The samples are prepared by Si Ion implantation into SiO_2

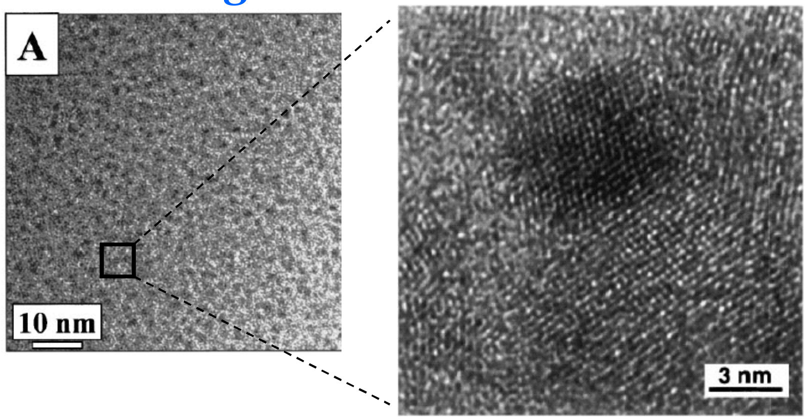
1 • Multi-energy implant (35-500 keV) yields uniform NC density

2 • Samples annealed @ 1100 C / 1 hr in Ar + H_2 to precipitate NC formation

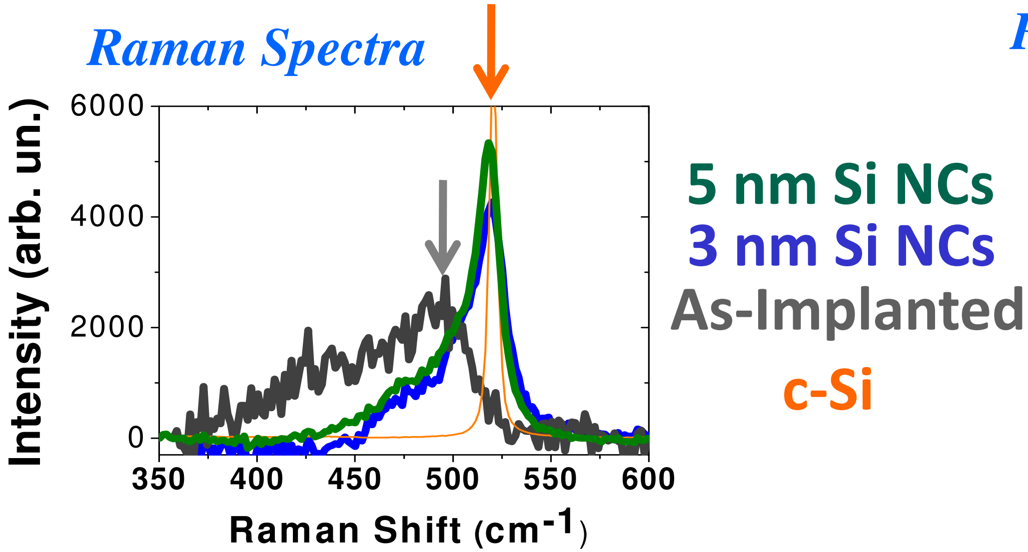
*C. W. White et al., NIM B **141**, 228 (1998) - ORNL*



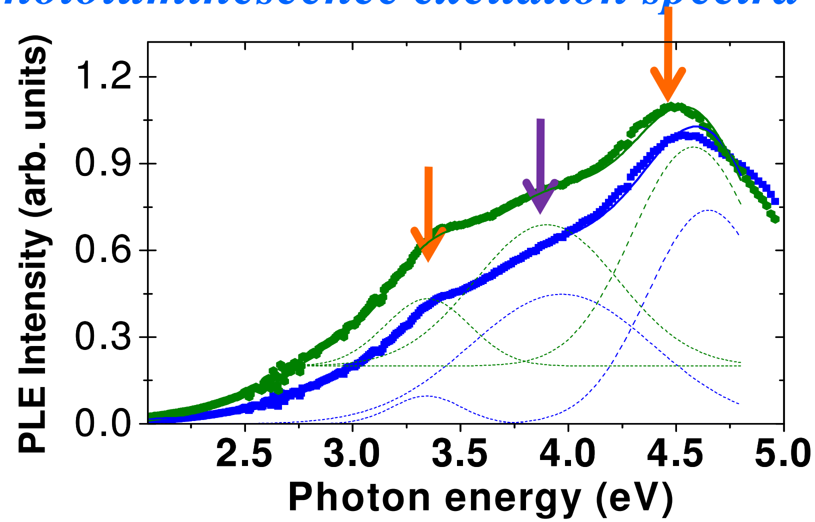
TEM Images



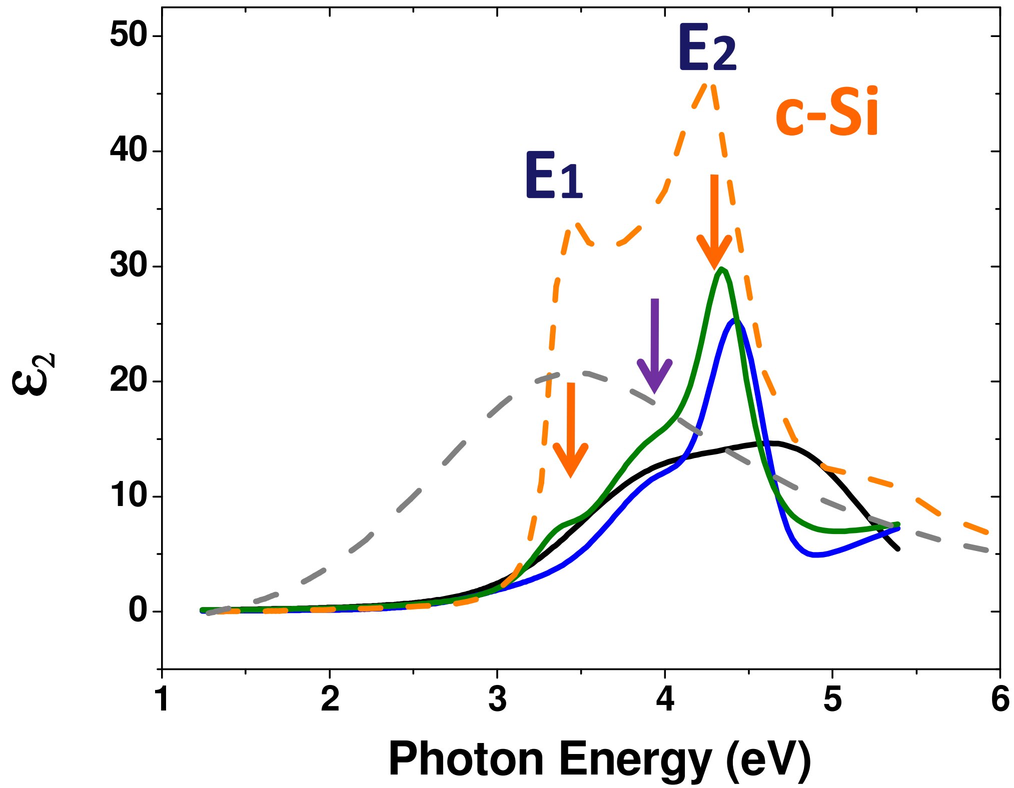
Raman Spectra



Photoluminescence excitation spectra



Spectroscopic elllipsometry (SE) shows modified c-Si E_1 and E_2 critical points in the Si NCs



5 nm and 3 nm Si NCs:

- Bulk CPs E_1 and E_2 preserved, with E_2 dominating the spectra and E_1 greatly suppressed
- Appearance of peak around 3.9 eV

As-Implanted:

- No bulk CPs E_1 and E_2
- Similar but blue-shifted shape to **a-Si**

- **SE spectra provide a comparison for SHG spectroscopy**
- **Measured $\varepsilon_{1,2}$ determine the Fresnel factors used in SHG analysis**

Cross-Polarized 2-Beam-SHG (XP2-SHG) enhances the signal from Si NCs by enhancing the field gradient

$$\vec{P}^{eff} = \Delta' \vec{E} \cdot \nabla \vec{E}$$

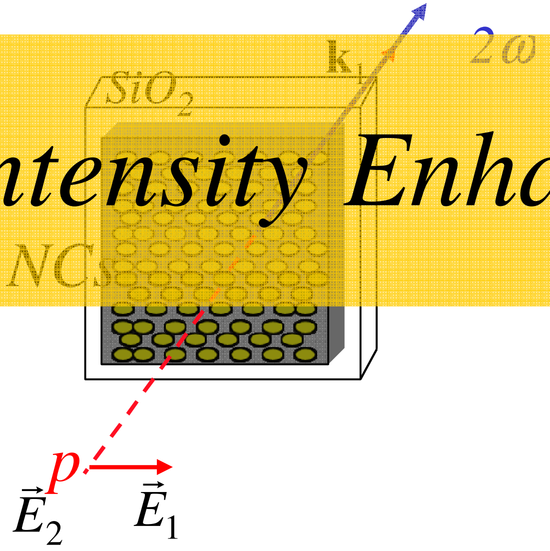
Single-beam SHG

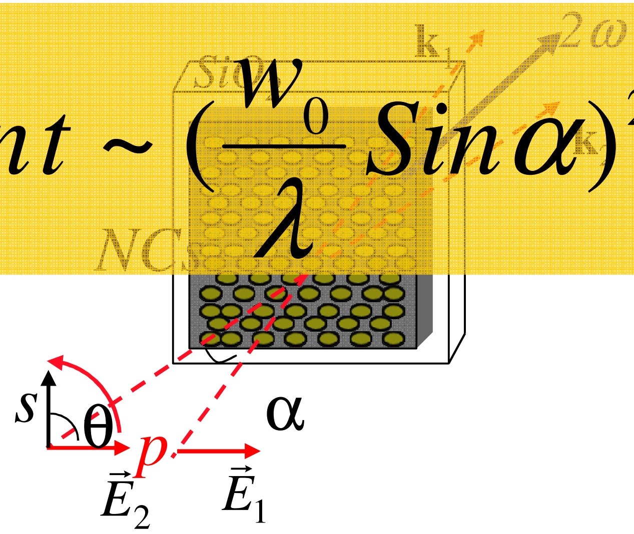
$$\vec{E} \cdot \nabla \vec{E} \sim \frac{E^2}{w_0}$$

XP2-SHG

$$\vec{E} \cdot \nabla \vec{E} \sim \frac{E_1 E_2}{\lambda} Sin\alpha$$

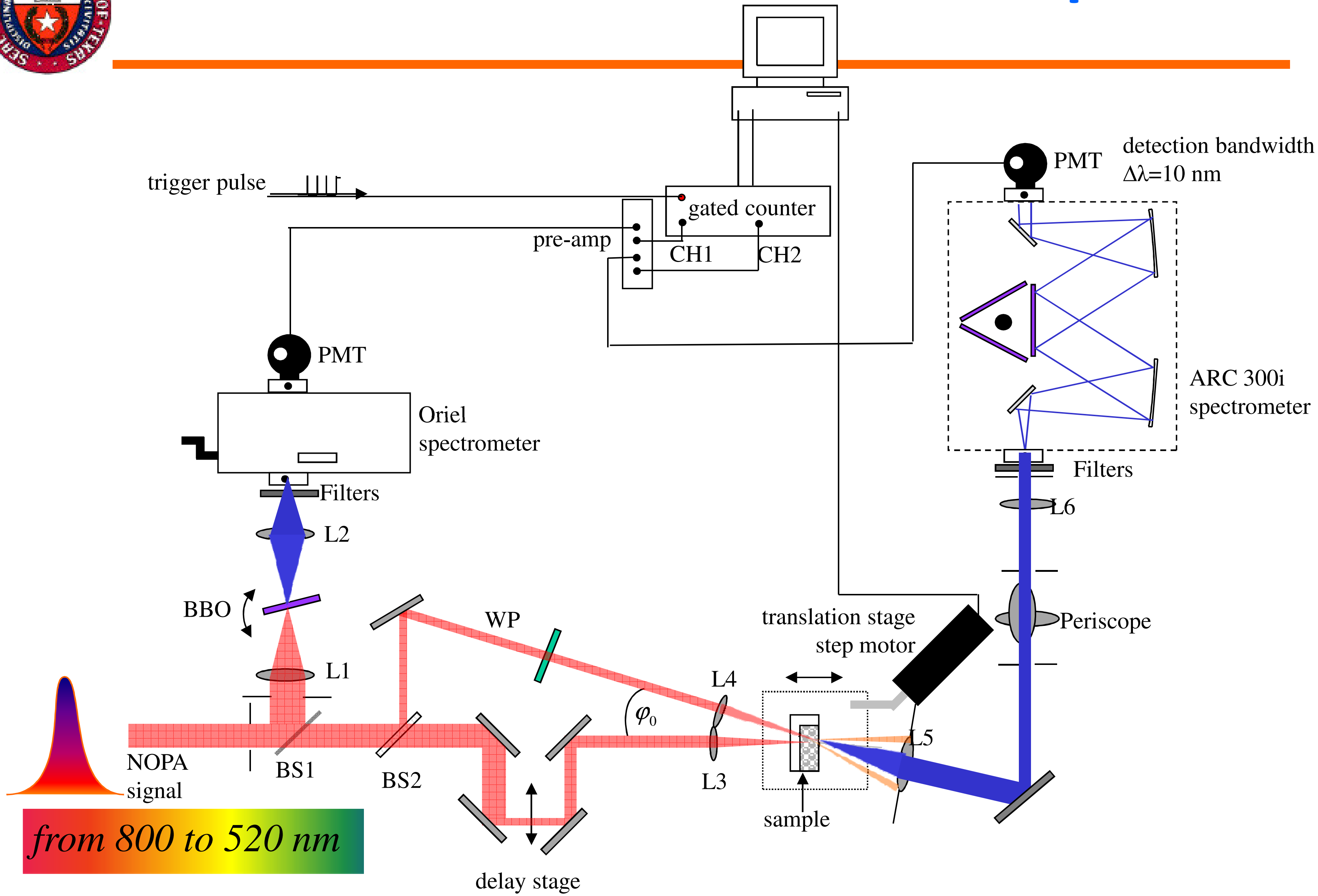
$$Intensity\ Enhancement \sim \left(\frac{w_0}{\lambda} Sin\alpha\right)^2$$



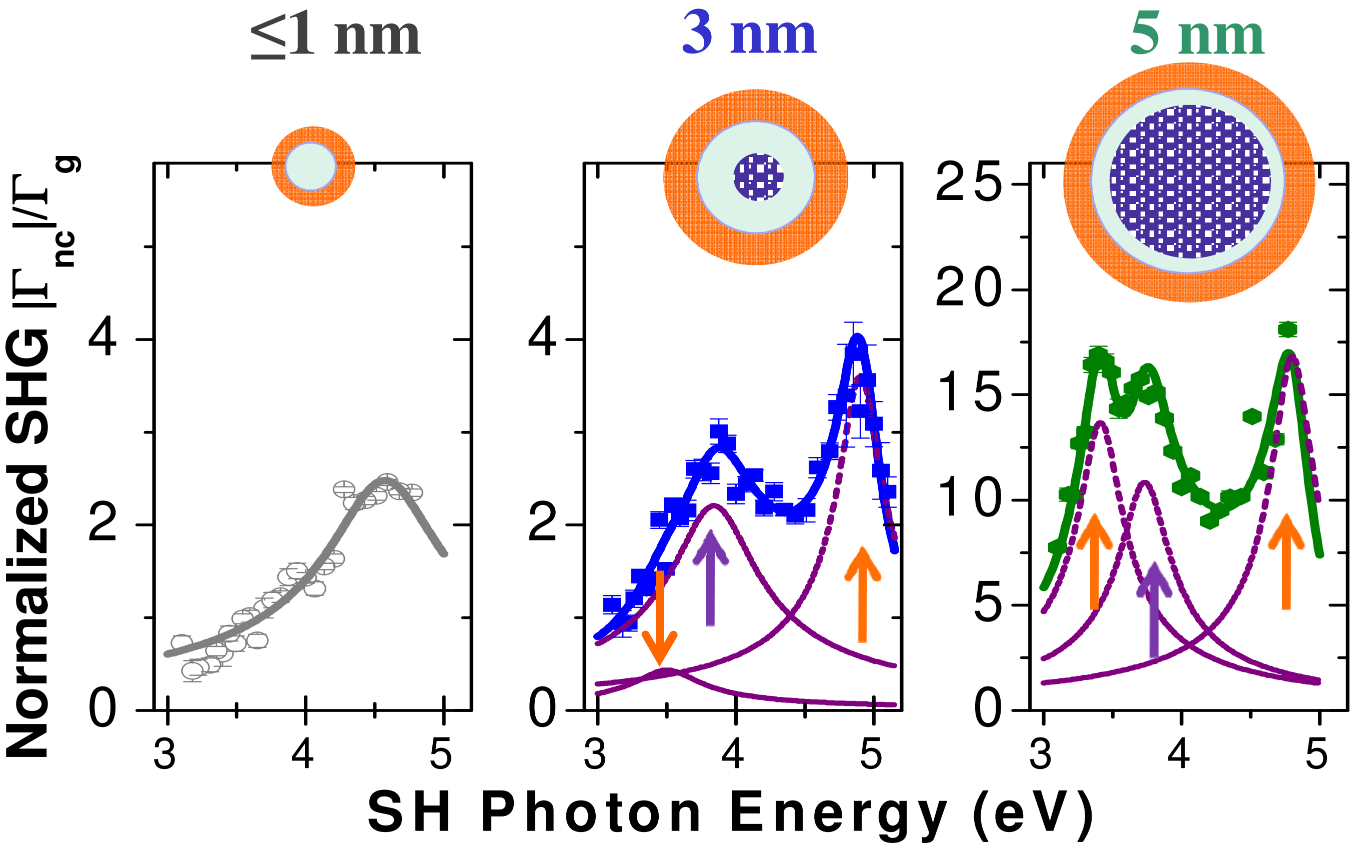


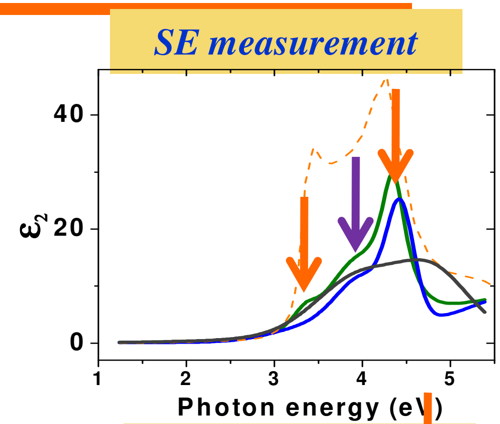
*L. Sun, et al, Opt. Lett., **30**, 2287 (2005)*
*P. Figliozzi, L. Sun, et al, Phy. Rev. Lett., **94**, 047401(2005)*

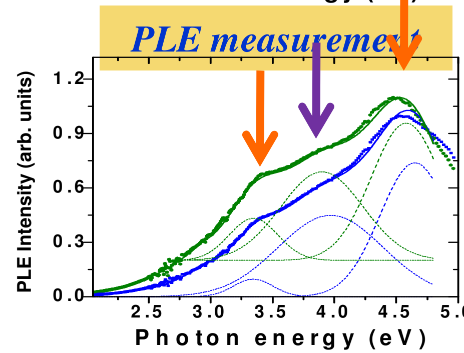
XP2-SHG Measurement Setup

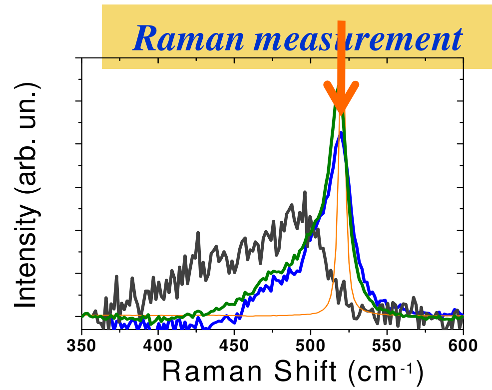


SHG spectra show strong interface resonance and modified c-Si critical points in the Si NCs







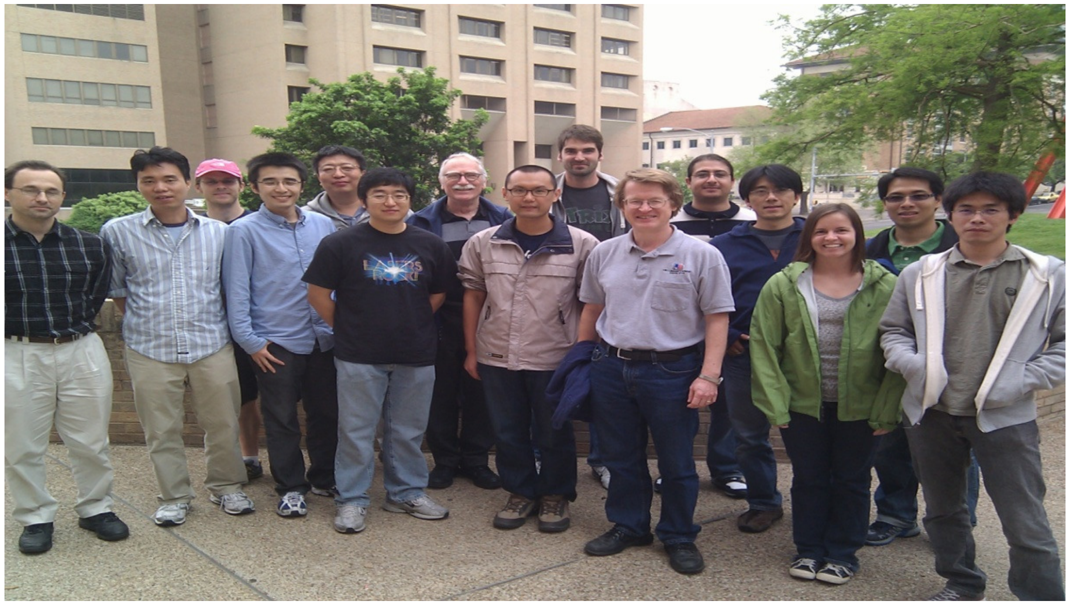


Quadrupolar SHG appears to be selectively sensitive to nano-interface structure (in close analogy to dipolar SHG of planar interfaces)

Conclusion

- **SHG, complemented with SE, PLE and Raman, has been applied to study Si NCs to help elucidate the unique structure of the NCs**
- **The unique sensitivity of SHG spectral structure and amplitude suggest SHG is uniquely sensitive to nano-interfacial structure**

Future directions:
1. Pump-probe XP2-SHG for dynamics study
2. Free-standing Si NCs



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