Enhanced Nonlinear Optical Response from Nano- and Micro-Scale Composite Materials

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with special thanks to:
Nick Lepeshkin, Aaron Schweinsberg, John Sipe
Giovanni Piredda, David D. Smith, and many others.

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The Promise of Nonlinear Optics

Nonlinear optical techniques hold great promise for applications including:

- **Photonic Devices**
- **Quantum Imaging**
- **Quantum Computing/Communications**
- **Optical Switching**
- **Optical Power Limiters**
- **All-Optical Image Processing**

But the lack of high-quality photonic material is often the chief limitation in implementing these ideas.

Composite Materials for Nonlinear Optics

Want large nonlinear response for applications in photonics

One specific goal:

Composite with $\chi^{(3)}$ exceeding those of constituents

Approaches:

- Nanocomposite materials
  Distance scale of mixing $<< \lambda$
  Enhanced NL response by local field effects

- Microcomposite materials (photonic crystals, etc.)
  Distance scale of mixing $\approx \lambda$
  Constructive interference increase E and NL response

Material Systems for Composite NLO Materials

All-dielectric composite materials

Minimum loss, but limited NL response

Metal-dielectric composite materials

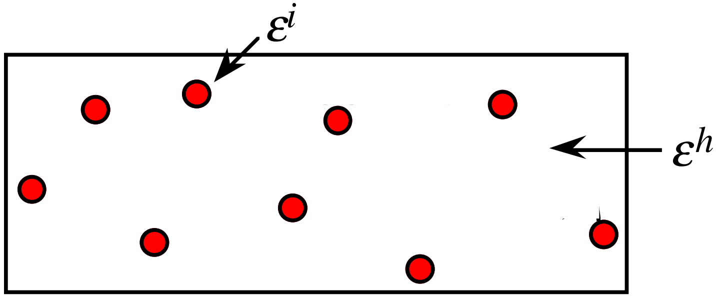
Larger loss, but larger NL response

Note that $\chi^{(3)}$ of gold $\approx 10^6$ $\chi^{(3)}$ of silica glass!

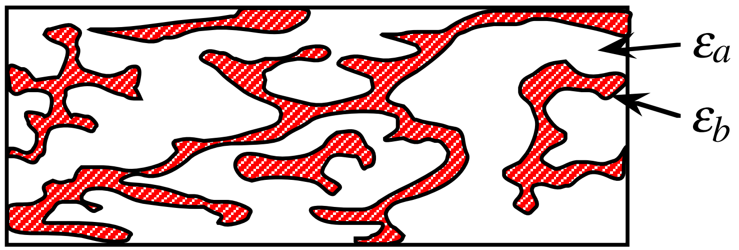
Also, metal-dielectric composites possess surface plasmon resonances, which can further enhance the NL response.

Nanocomposite Materials for Nonlinear Optics

- Maxwell Garnett

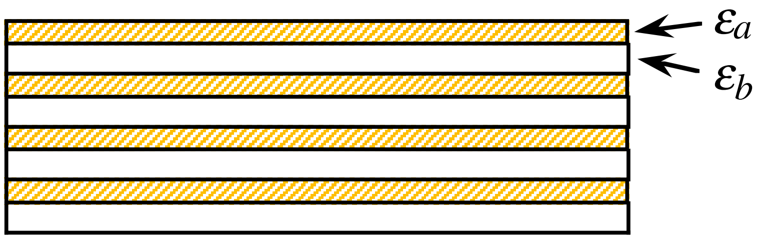


- Bruggeman (interdispersed)



- Fractal Structure

- Layered



scale size of inhomogeneity << optical wavelength

Gold-Doped Glass: A Maxwell-Garnett Composite

Red Glass Caraffe
Nurenberg, ca. 1700

Huelsmann Museum, Bielefeld

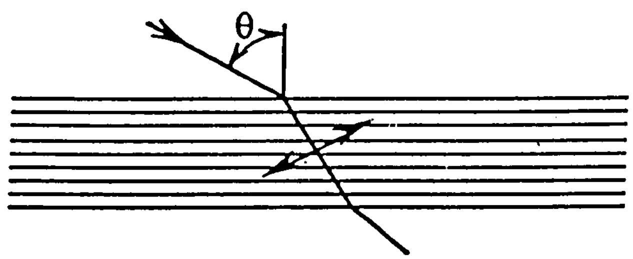
Developmental Glass, Corning Inc.

gold volume fraction approximately 10^{-6}
gold particles approximately 10 nm diameter

- Composite materials can possess properties very different from those of their constituents.
- Red color is because the material absorbs very strong in the blue, at the surface plasmon frequency

Demonstration of Enhanced NLO Response

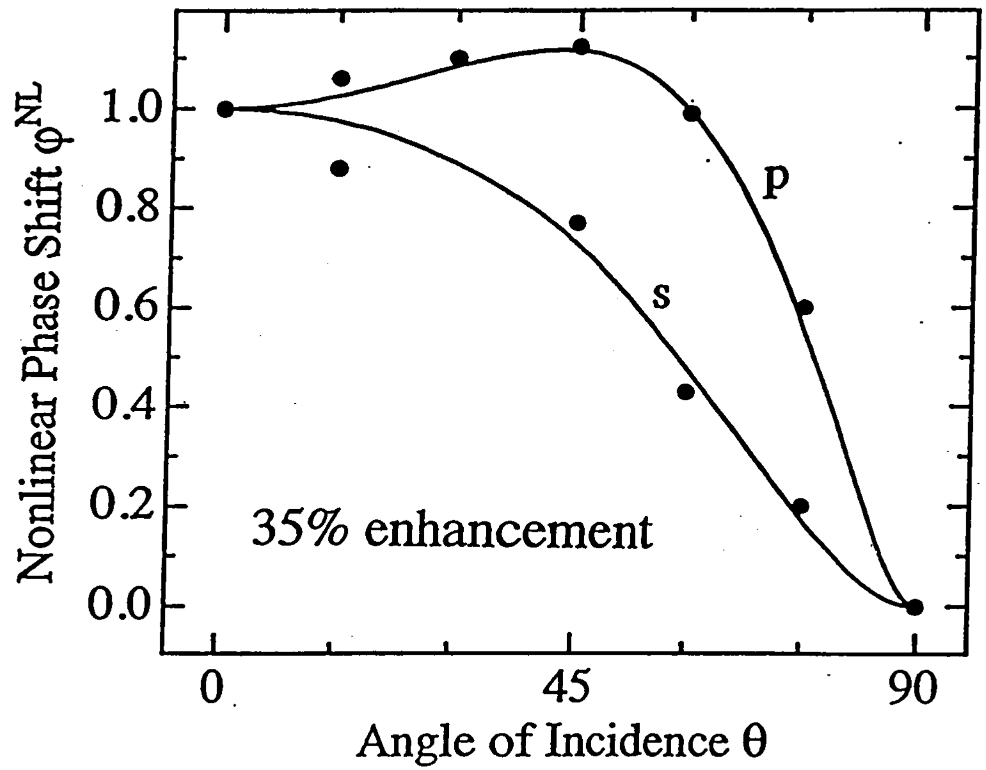
- Alternating layers of TiO_2 and the conjugated polymer PBZT.



$\nabla \cdot \mathbf{D} = 0$ implies that $(\varepsilon \mathbf{E})_\perp$ is continuous.

Thus field is concentrated in *lower* index material.

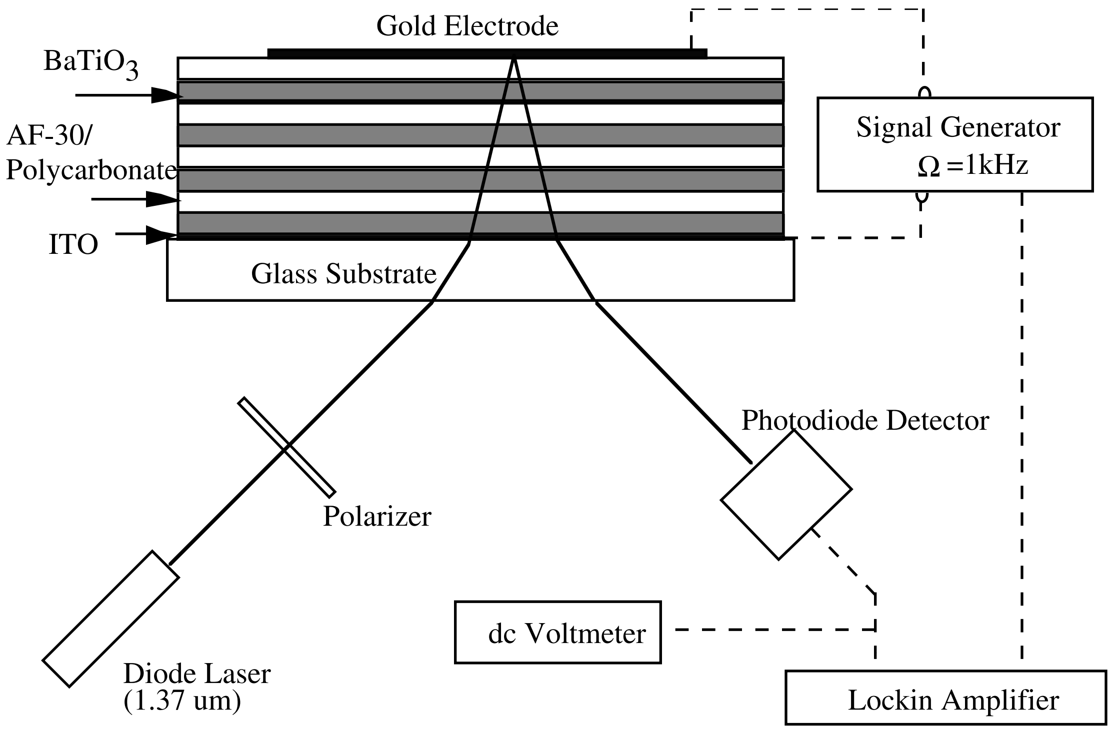
- Measure NL phase shift as a function of angle of incidence



Fischer, Boyd, Gehr, Jenekhe, Osaheni, Sipe, and Weller-Brophy, Phys. Rev. Lett. 74, 1871, 1995.

Gehr, Fischer, Boyd, and Sipe, Phys. Rev. A 53, 2792 1996.

Enhanced EO Response of Layered Composite Materials



$$\chi_{ijkl}^{(eff)}(\omega';\omega,\Omega_1,\Omega_2) = f_a \left[\frac{\varepsilon_{eff}(\omega')}{\varepsilon_a(\omega')}\right]\left[\frac{\varepsilon_{eff}(\omega)}{\varepsilon_a(\omega)}\right]\left[\frac{\varepsilon_{eff}(\Omega_1)}{\varepsilon_a(\Omega_1)}\right]\left[\frac{\varepsilon_{eff}(\Omega_2)}{\varepsilon_a(\Omega_2)}\right]\chi_{ijkl}^{(a)}(\omega';\omega,\Omega_1,\Omega_2)$$

- AF-30 (10%) in polycarbonate (spin coated)
  n=1.58 ε(dc) = 2.9
- barium titante (rf sputtered)
  n=1.98 ε(dc) = 15

$$\chi_{zzzz}^{(3)} = (3.2 + 0.2i)\times 10^{-21}\ (m/V)^2 \pm 25\%$$

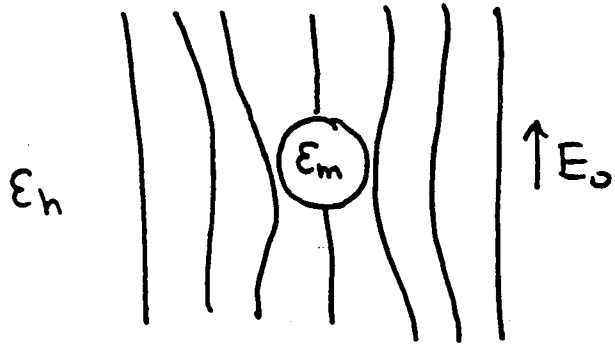
$$\approx 3.2\ \chi_{zzzz}^{(3)}(\text{AF-30 / polycarbonate})$$

3.2 times enhancement in agreement with theory

R. L. Nelson, R. W. Boyd, Appl. Phys. Lett. 74, 2417, 1999.

Metal / Dielectric Composites

Very large local field effects



$$E_{in} = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h} E_0$$

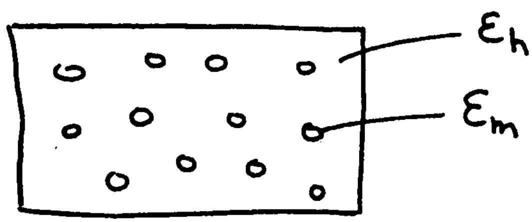
$$\equiv \mathcal{L} E_0$$

(ε_m is negative !)

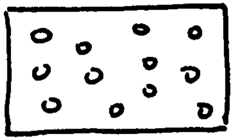
At resonance

$$\mathcal{L} = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h} \longrightarrow \frac{3\varepsilon_h}{i\,\varepsilon_m''} \approx (3 \text{ to } 30)\, i$$

$$\chi^{(3)}_{eff} = f\, \mathcal{L}^2 |\mathcal{L}|^2 \chi^{(3)}_m + (1-f)\, \chi^{(3)}_h$$



Counterintuitive Consequence of Local Field Effects



gold nanoparticles in a liquid dye solution (HITCI)

Both constituents are reverse saturable absorbers $\Rightarrow$ $\text{Im}\,\chi^{(3)} > 0$

Effective NL susceptibility of composite

$$\chi^{(3)}_{eff} = f\, L^2 |L|^2 \chi^{(3)}_{Au} + (1-f)\, \chi^{(3)}_{dye\ sol'n}$$

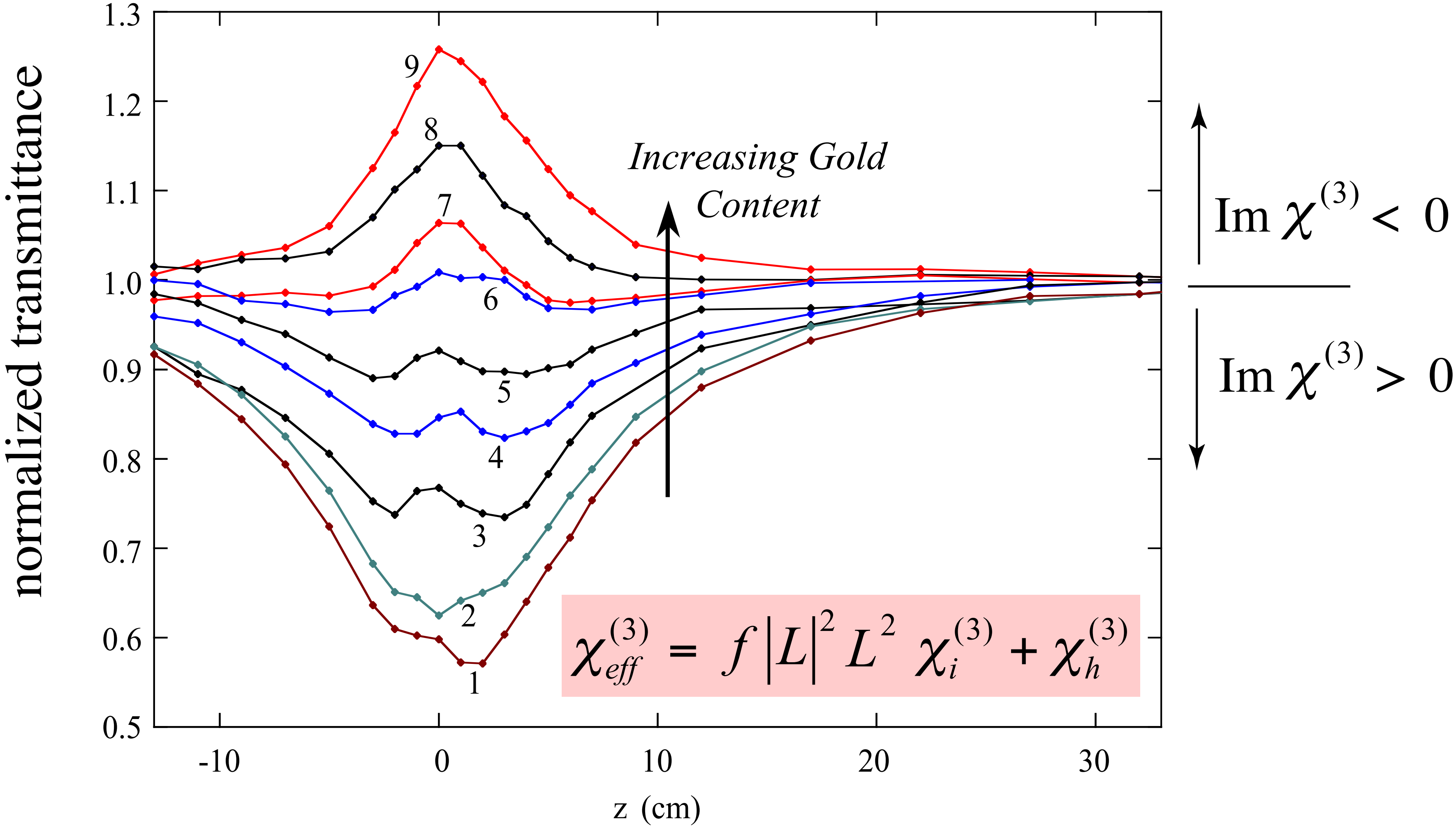
$$L = \frac{3\varepsilon_h}{\varepsilon_m + 2\varepsilon_h} = \text{pure imaginary at resonance!}$$

A cancellation of the two contributions to $\chi^{(3)}$ can occur, even though they have same sign.

Counterintuitive Consequence of Local Field Effects

Cancellation of two contributions that have the same sign

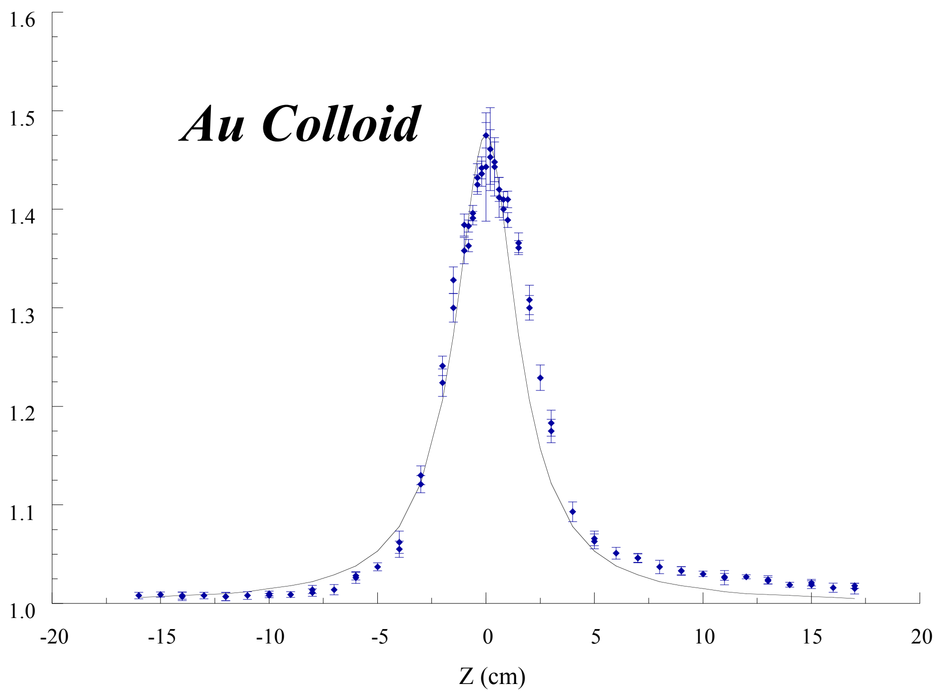
Gold nanoparticles in a saturable absorber dye solution (13 μM HITCI)

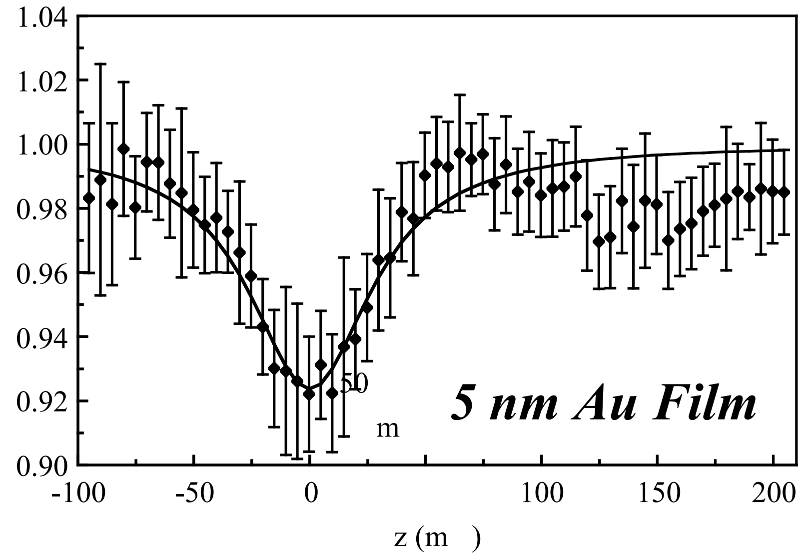


D.D. Smith, G. Fischer, R.W. Boyd, D.A.Gregory, *JOSA B* **14**, 1625, 1997.

Comparison of Bulk and Colloidal Gold

Open Aperture Z-Scans of Gold Colloid and Au film at 532nm

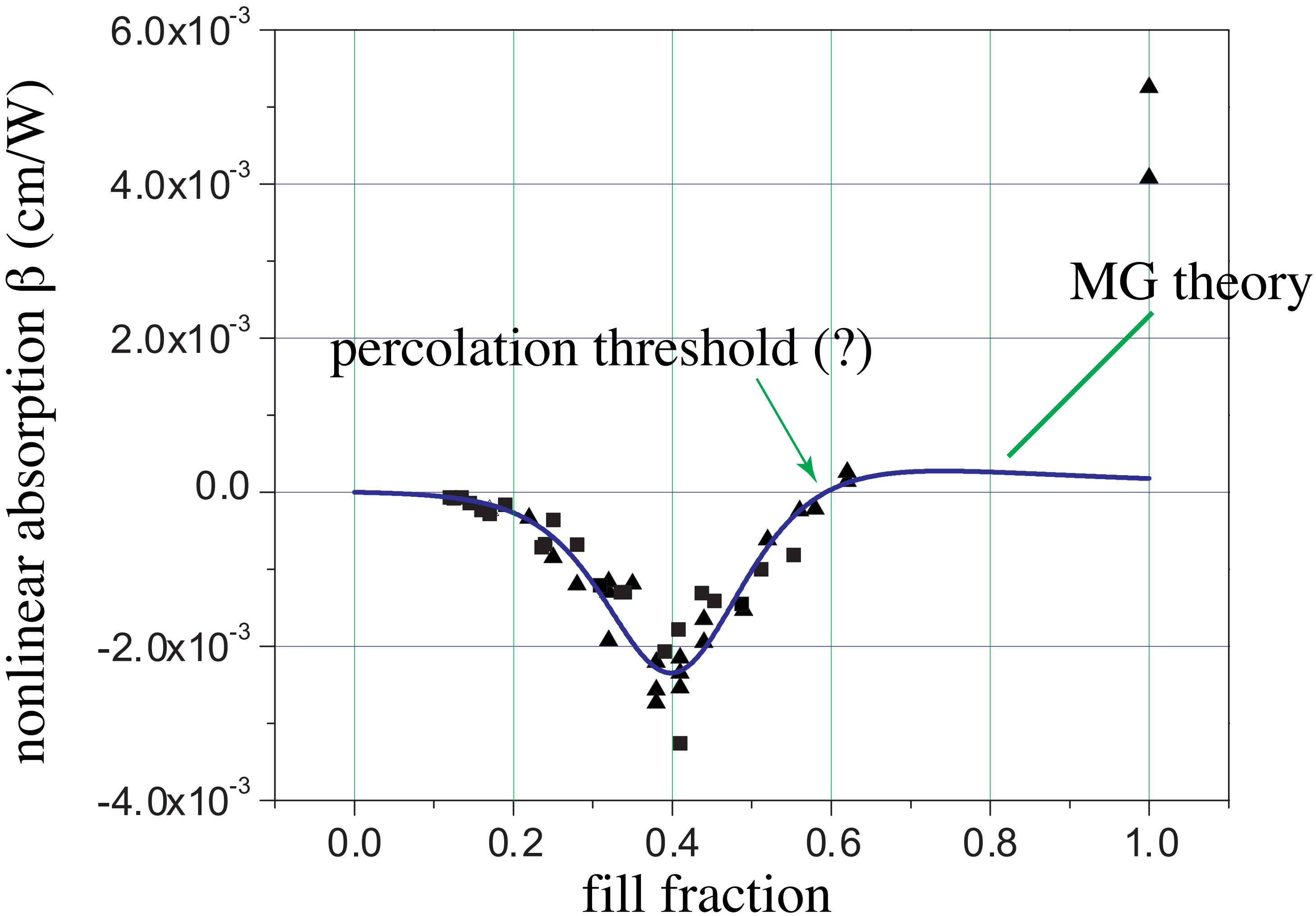


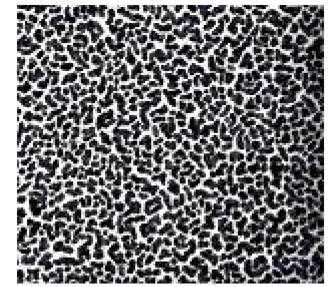


- ➢ Nonlinearities possess opposite sign!

Nonlinear Optical Response of Semicontinuous Metal Films

Measure nonlinear response as function of gold fill fraction



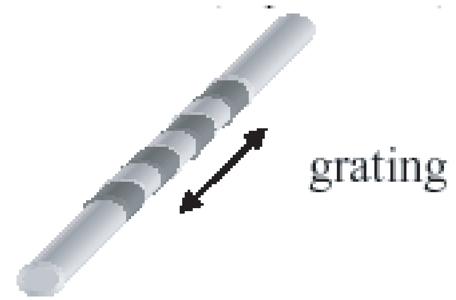


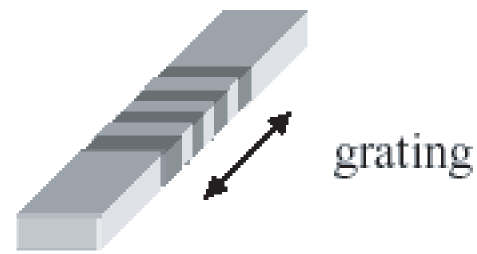
(with D. D. Smith and G. Piredda)

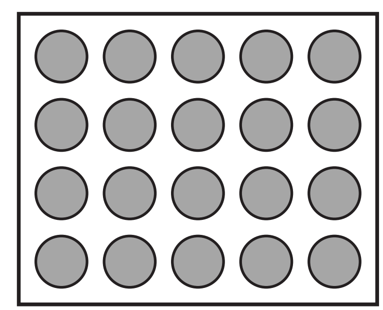
Artificial Materials for Nonlinear Optics

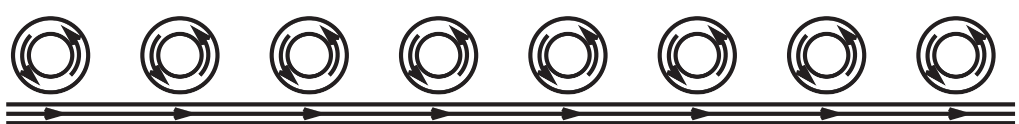
Artifical materials can produce
- Large nonlinear optical response
- Large dispersive effects

Examples
- Fiber/waveguide Bragg gratings
- PBG materials
- CROW devices (Yariv et al.)
- SCISSOR devices

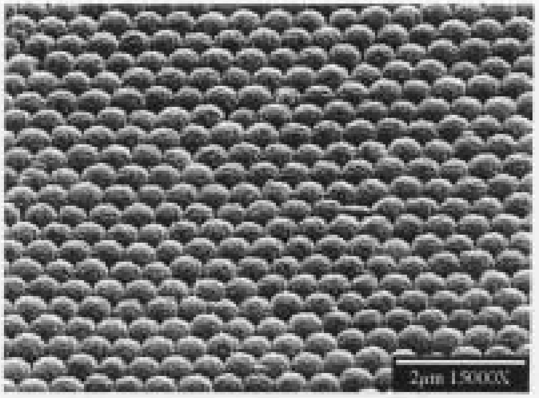






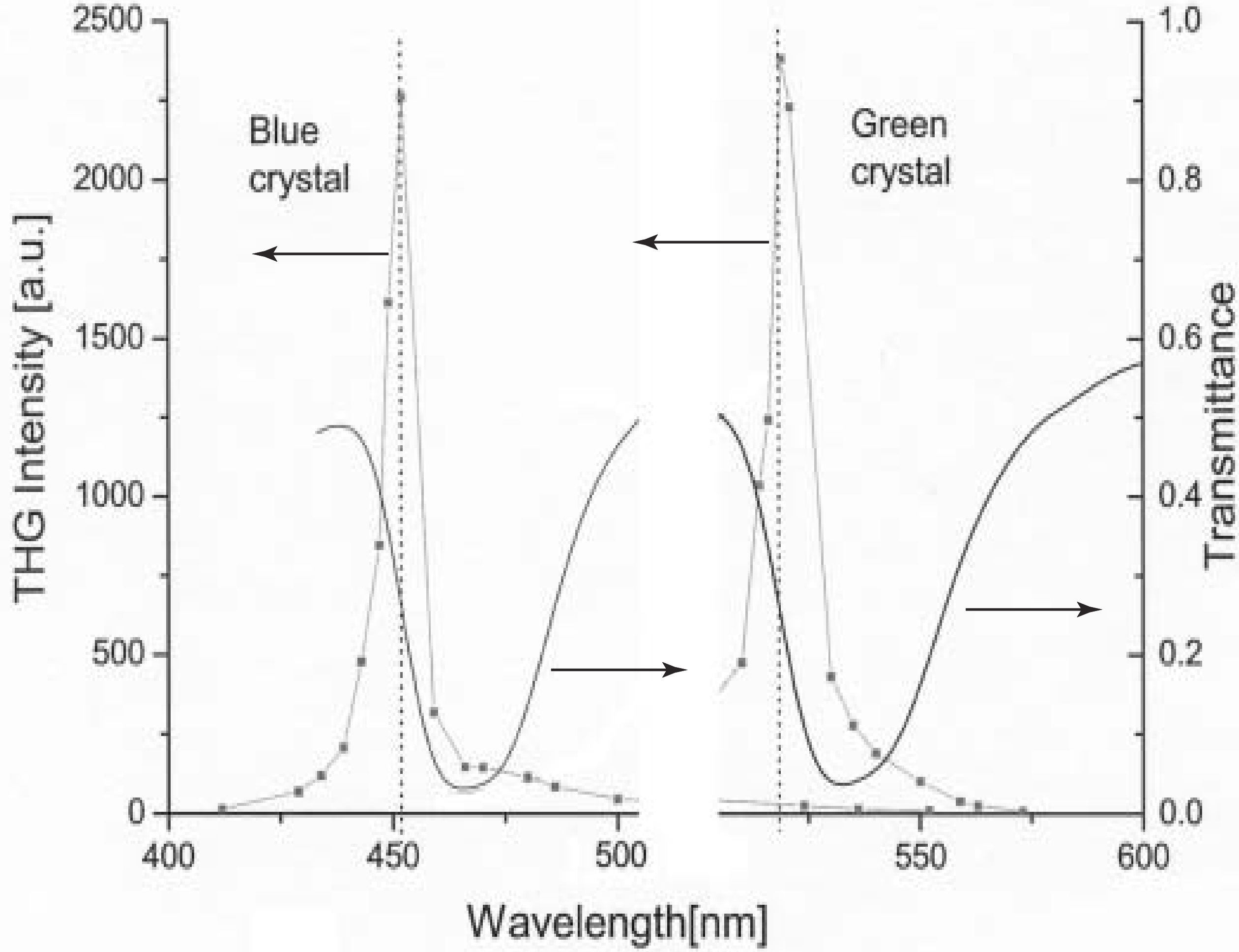


Third-Harmonic Generation in a 3D Photonic Crystal



polystyrene photonic crystal

Direct THG visible by eye!

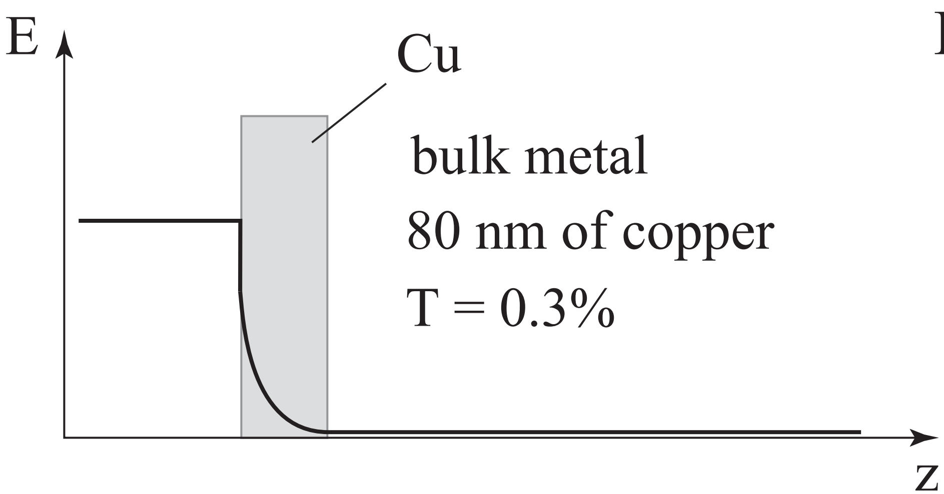


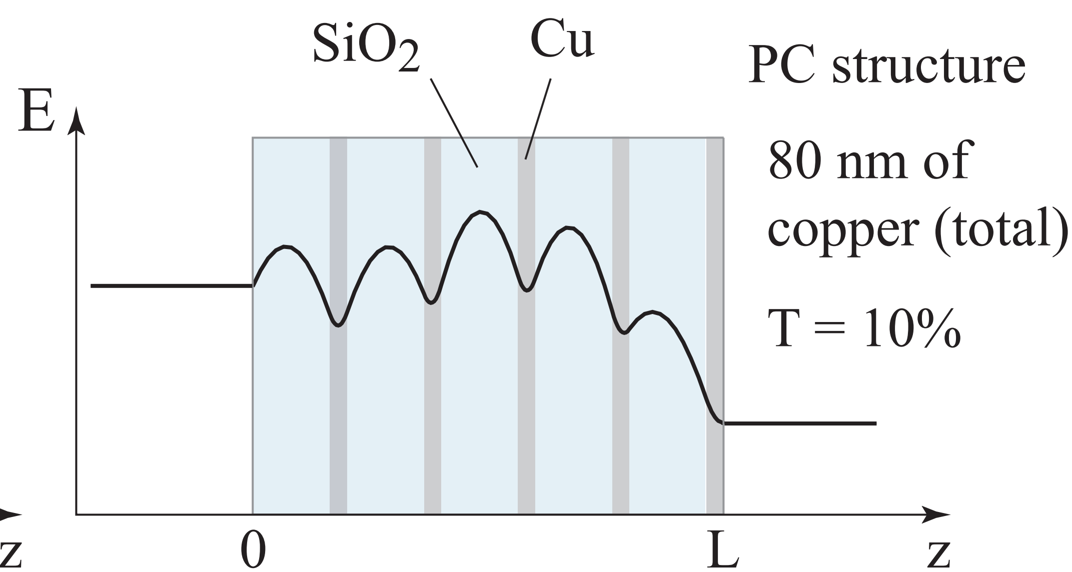
Phase matching provided by PBG structure.

P. P. Markowicz, V. K. S. Hsiao, H. Tiryaki, A. N. Cartwright, P. N. Prasad, K. Dolgaleva, N. N. Lepeshkin, and R. W. Boyd, Appl. Phys. Lett. 87, 051102 (2005)

Accessing the Optical Nonlinearity of Metals with Metal-Dielectric Photonic Crystal Structures

- Metals have very large optical nonlinearities but low transmission
- Low transmission is because metals are highly reflecting (not because they are absorbing!)
- Solution: construct metal-dielectric photonic crystal structure (linear properties studied earlier by Bloemer and Scalora)

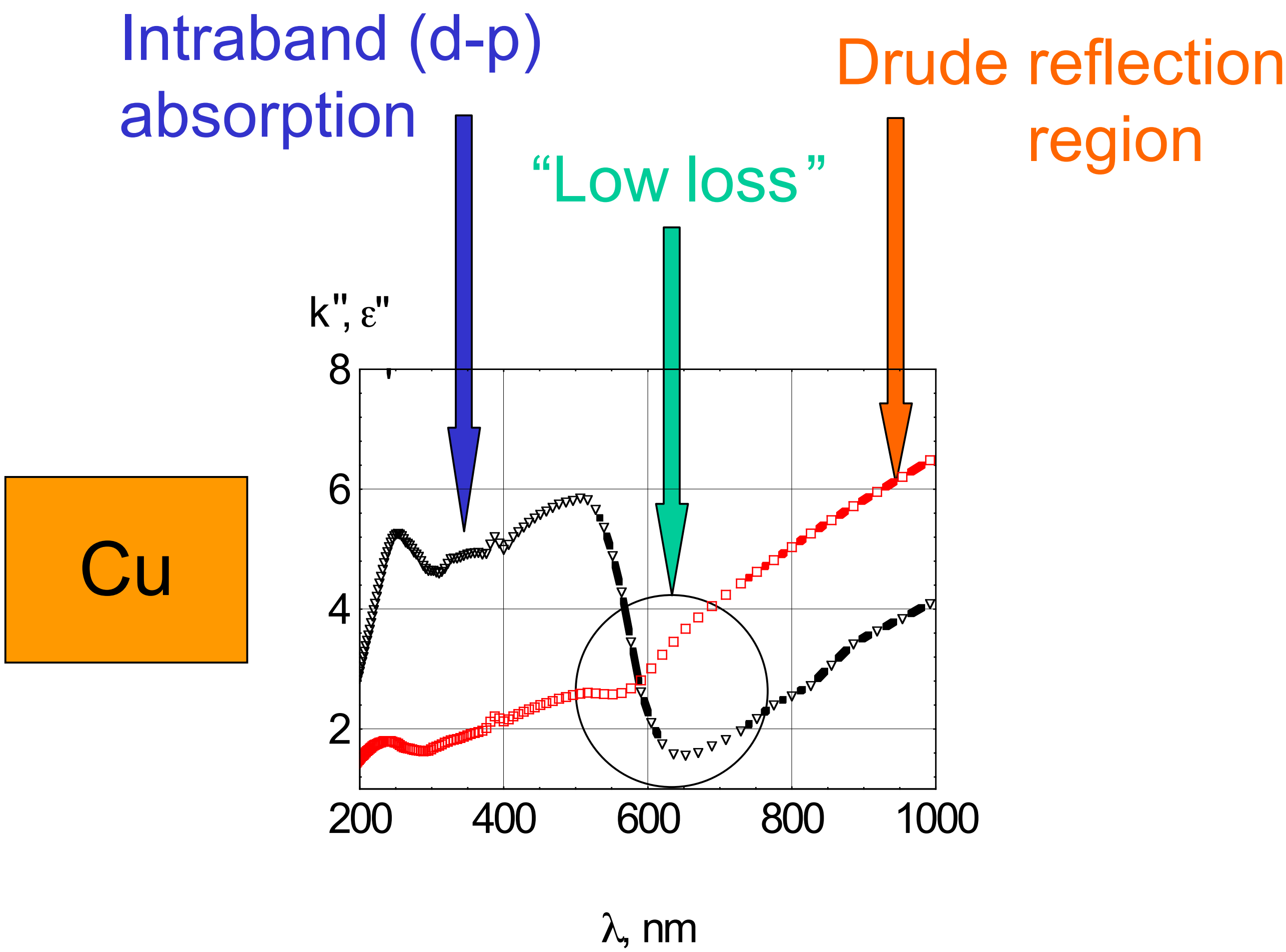




Greater than 10 times enhancement of NLO response is predicted!

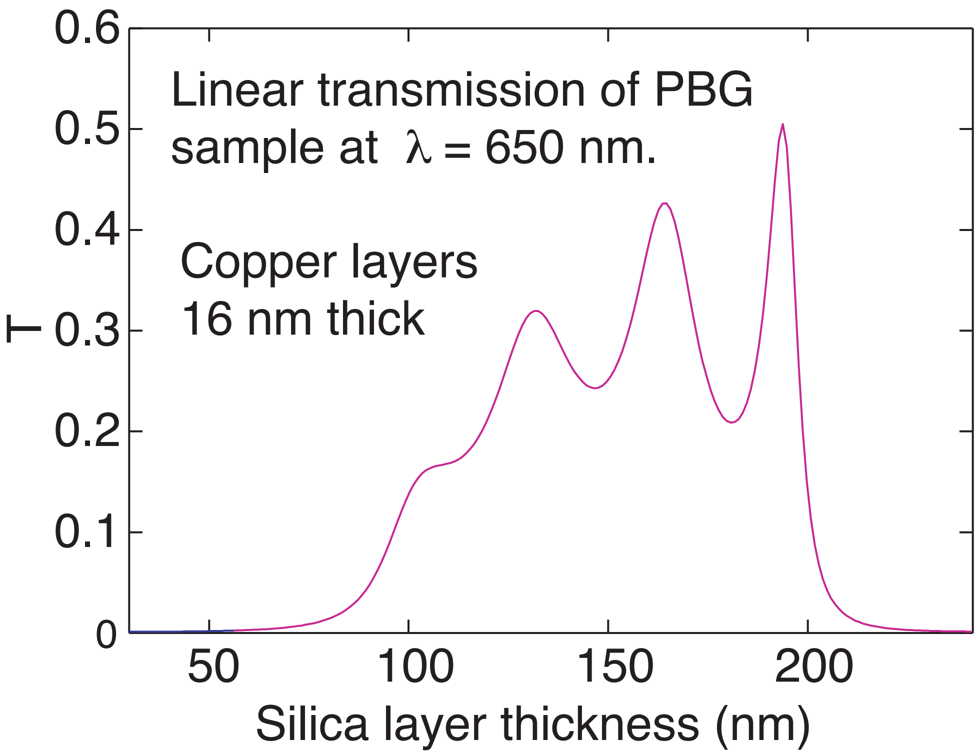
R.S. Bennink, Y.K. Yoon, R.W. Boyd, and J. E. Sipe, Opt. Lett. 24, 1416, 1999.

“Loss” mechanisms in copper

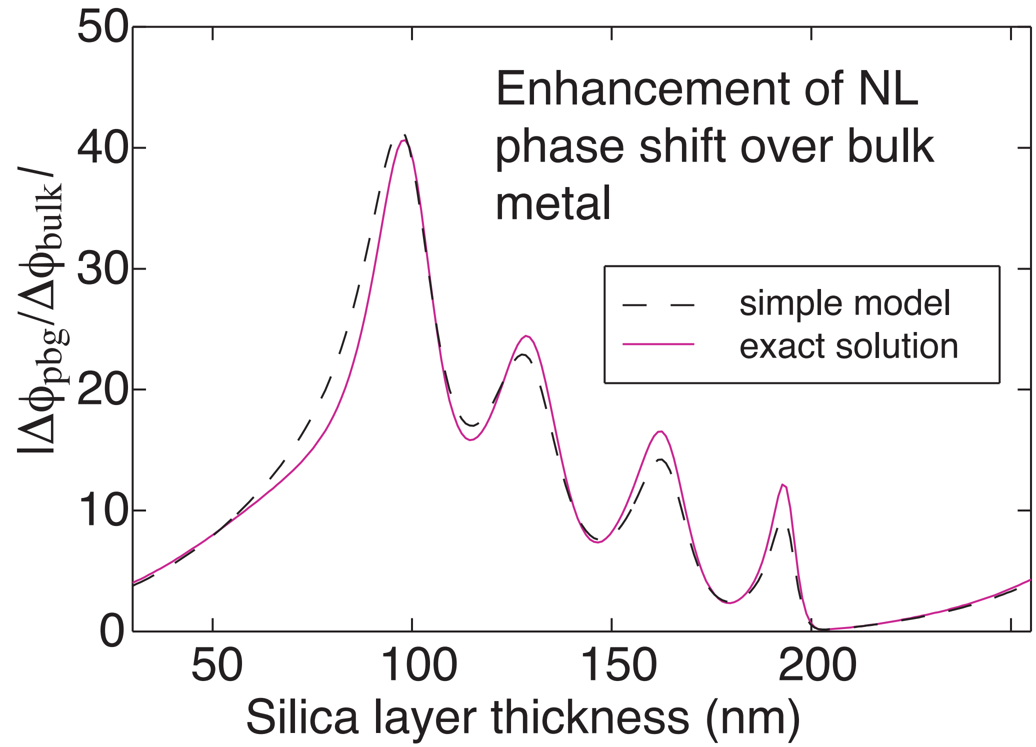


Accessing the Optical Nonlinearity of Metals with Metal-Dielectric Photonic Crystal Structures

- Metal-dielectric structures can have high transmission.



- And produce enhanced nonlinear phase shifts!

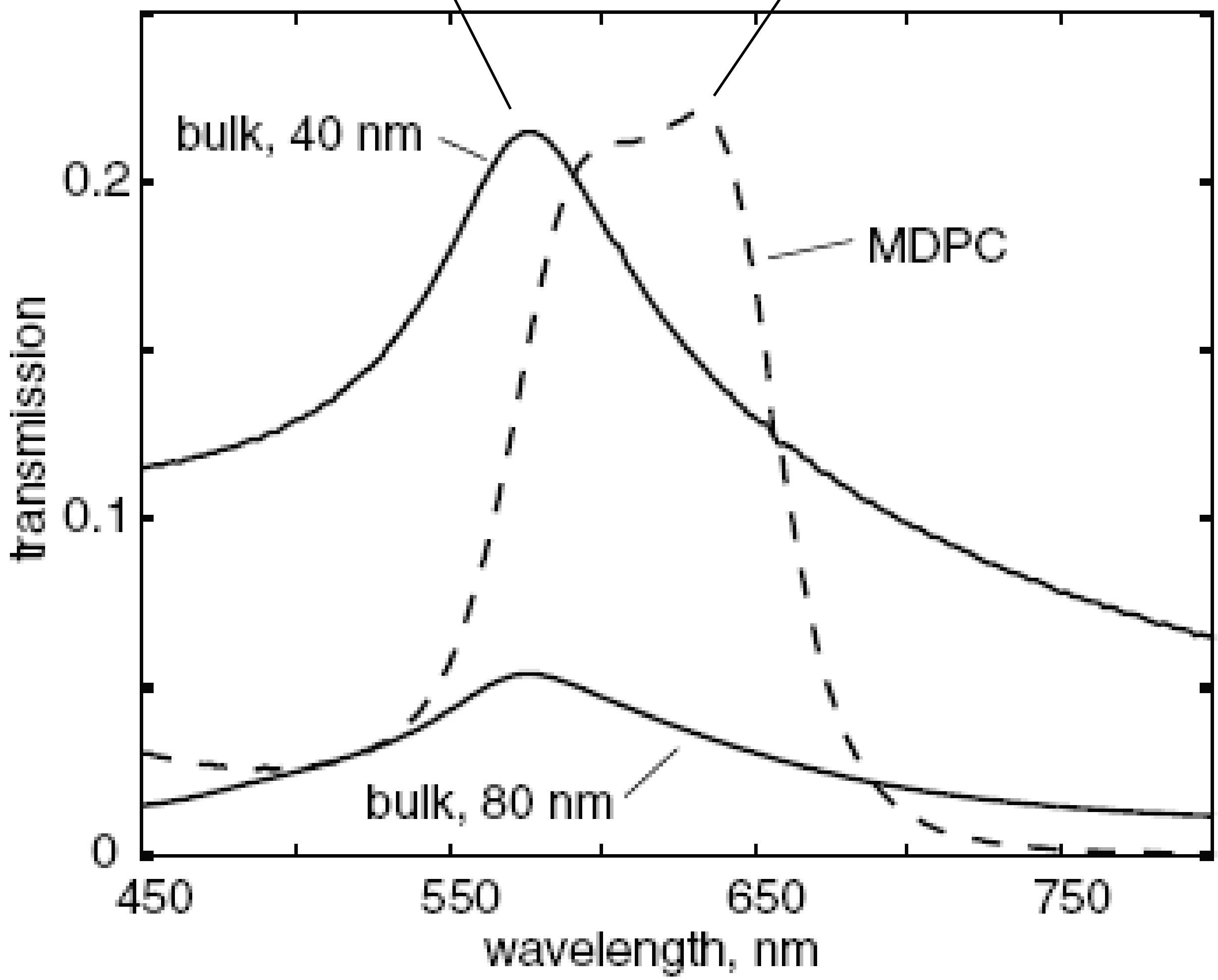


- The imaginary part of $\chi^{(3)}$ produces a nonlinear phase shift! (And the real part of $\chi^{(3)}$ leads to nonlinear transmission!)

Linear Transmittance of Samples

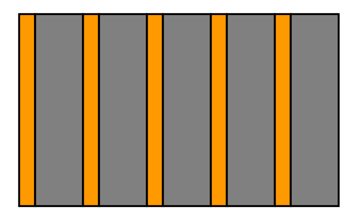
Material (interband) feature

Structural (M/D PC) feature



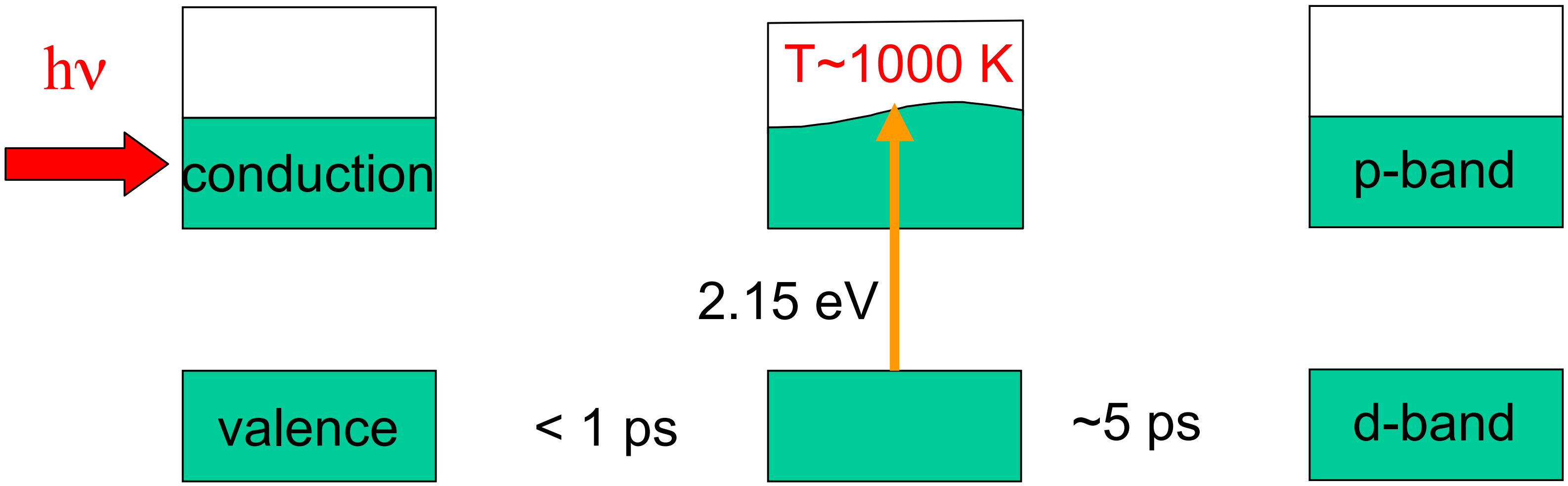
Cu: 40 nm film

M/D PC: Cu / silica



5x16/98 nm
(80 nm total Cu)

Mechanism of nonlinear response: “Fermi smearing”



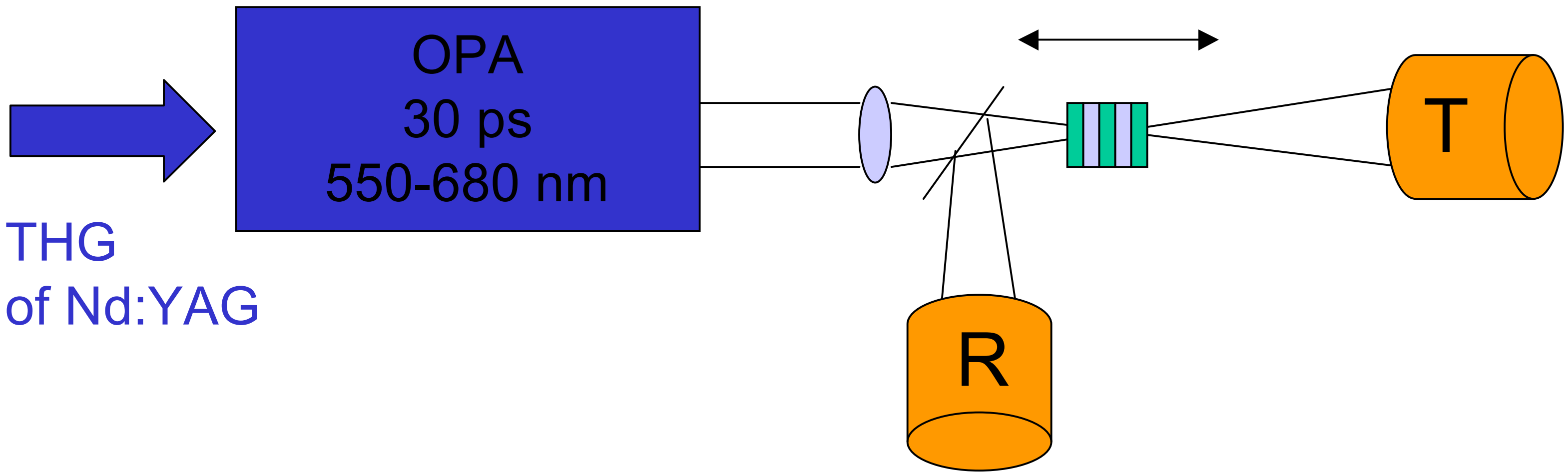
$$\Delta T \rightarrow \Delta\varepsilon(E_{IB}) \rightarrow \text{change in optical properties}$$

Near the interband absorption edge, “Fermi smearing” is the dominant nonlinear process

$\chi^{(3)}$ is largely imaginary

G. L. Eesley, Phys. Rev. B33, 2144 (1986)
H. E. Elsayed-Ali et al. Phys. Rev. Lett. 58, 1212 (1987)

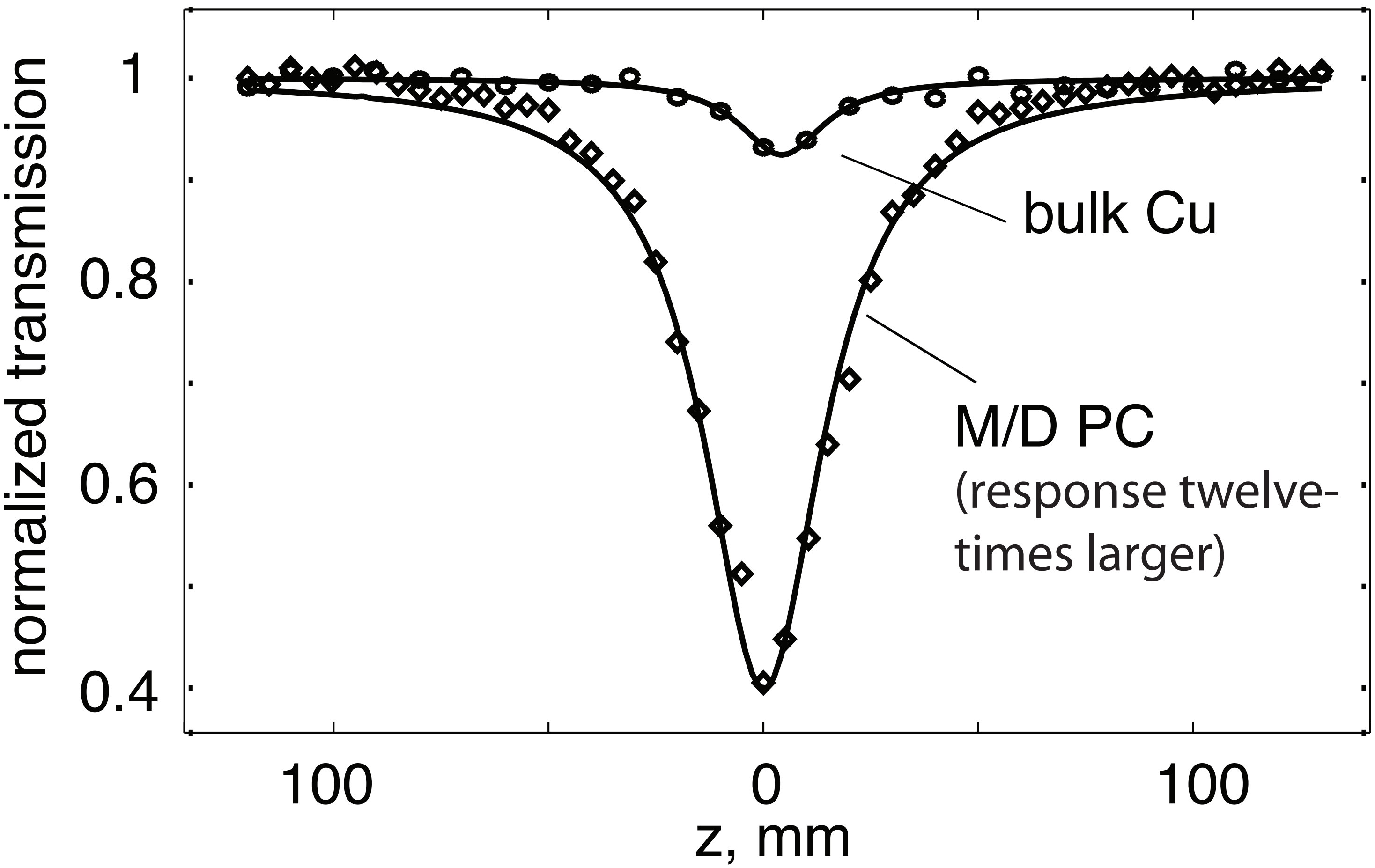
Reflection/Transmission Z-Scan



Pulse energy ~ 1m J
I = 100 MW/cm^2

$$\frac{\Delta R}{R}, \frac{\Delta T}{T} \rightarrow \Delta\varepsilon' + \Delta\varepsilon''$$

Z-Scan Comparison of M/D PC and Bulk Sample



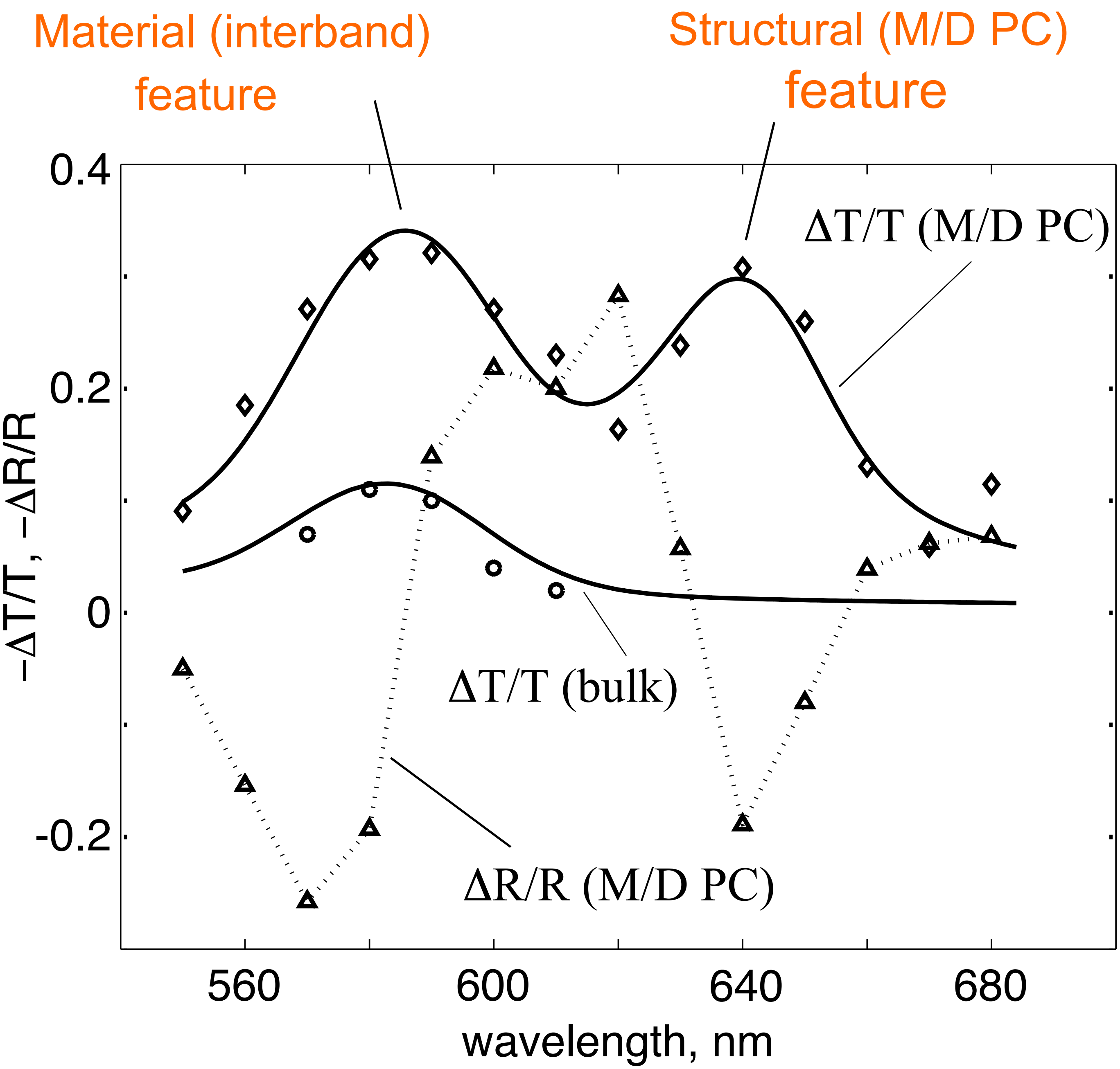
I = 500 MW/cm^2

λ = 650 nm

- We observe a large NL change in transmission
- But there is no measurable NL phase shift for either sample

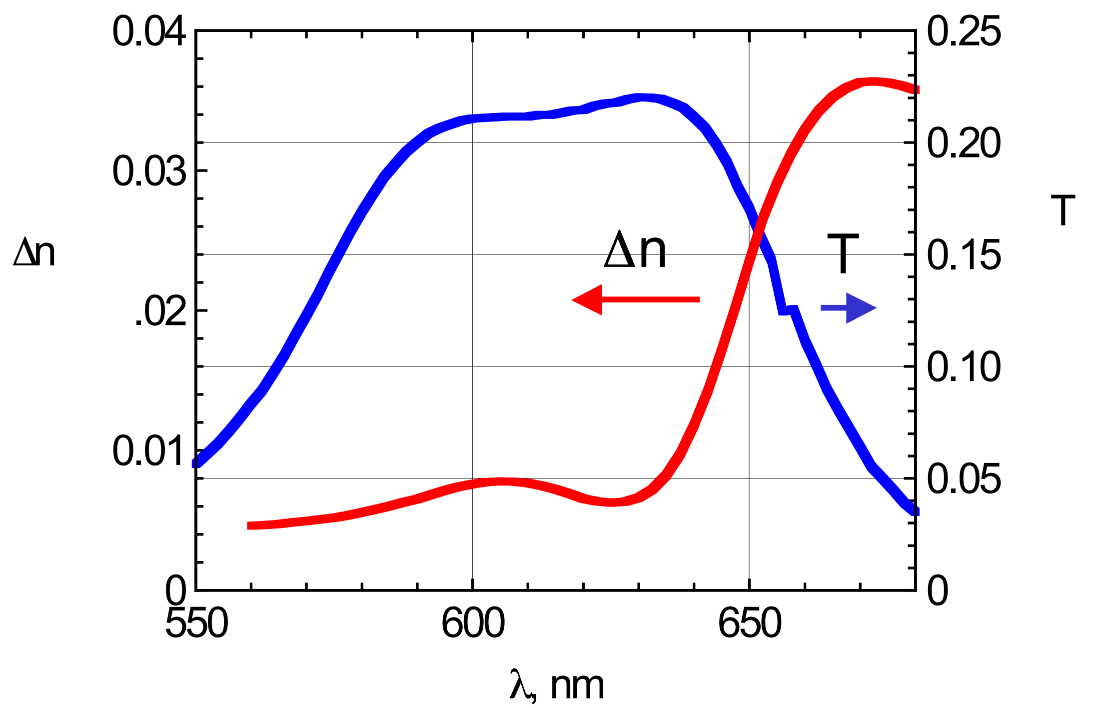
Lepeshkin, Schweinsberg, Piredda, Bennink, Boyd, Phys. Rev. Lett. 93 123902 (2004).

Nonlinear Transmission and Reflectance



Nonlinear phase shift of PC (numerical simulations)

$$\Delta\varepsilon = 0.1i \rightarrow \Delta n$$

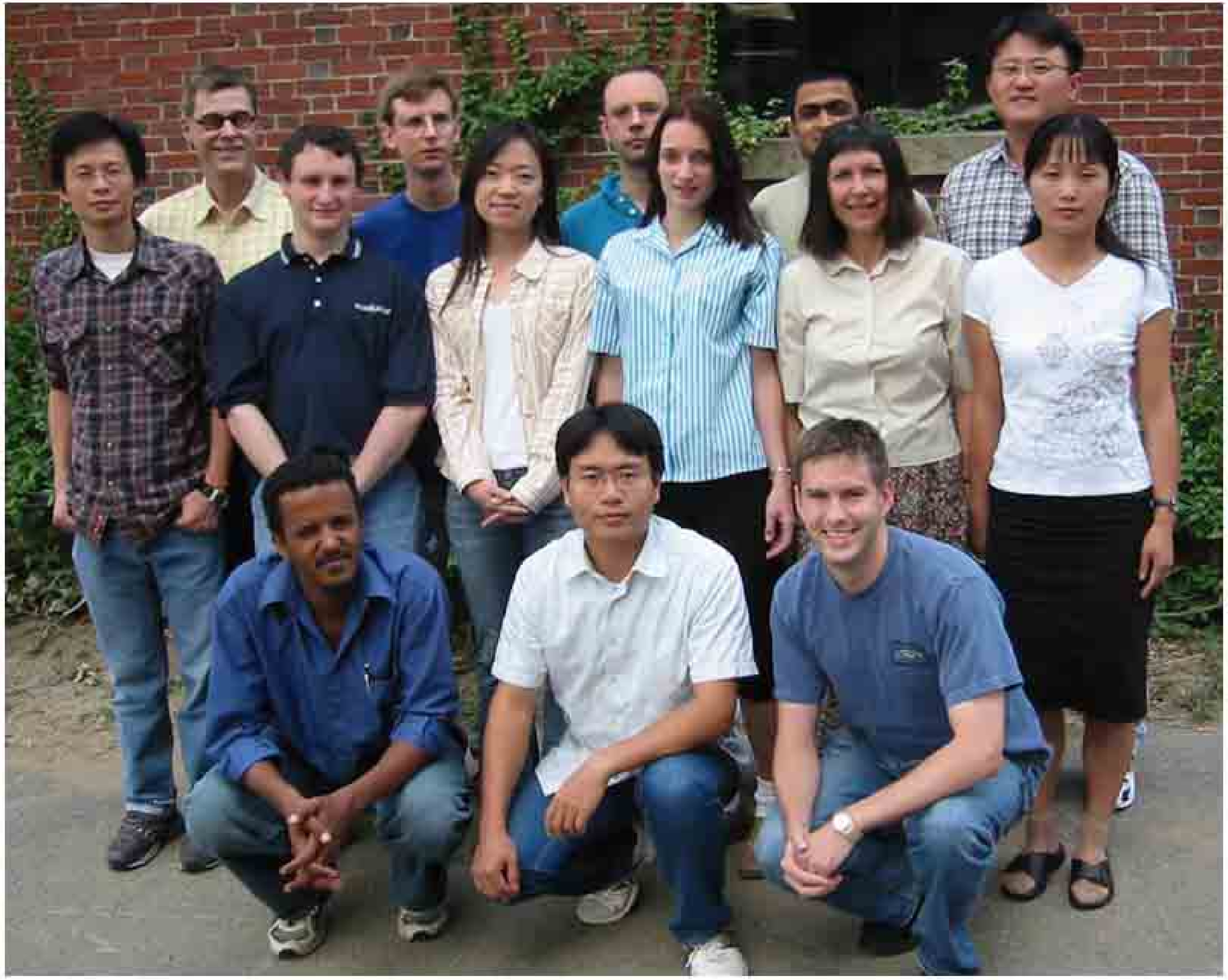


- Appreciable Δn occurs only outside of transparency window
- Next step: design and fabricate new structure

Conclusions

- Both nano-scale and microscale structuring can lead to enhanced nonlinear optical effects
- Influence of nano-scale structuring can be understood in terms of local field effects
- Nano-scale structuring can lead to enhancement (layered results) or cancellation (dye/colloid) of NLO response
- Influence of microscale structuring can be understood in terms of properties of photonic crystals
- Dispersion induced by photonic crystal can lead to new phase-matching effects
- Metal / dielectric photonic crystals can be designed to allow access to the large nonlinearity of metals

Special Thanks to My Students and Research Associates



Thank you for your attention!

Approaches to the Development of Improved NLO Materials

- New chemical compounds
- Quantum coherence (EIT, etc.)
- Composite Materials:

  (a) Microstructured Materials, e.g.
  Photonic Bandgap Materials,
  Quasi-Phasematched Materials, etc

  (b) Nanocomposite Materials

These approaches are not incompatible and in fact can be exploited synergistically!