Metal-dielectric composites as nonlinear optical materials

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Composite Materials for Nonlinear Optics

Want large nonlinear response for applications in photonics

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!

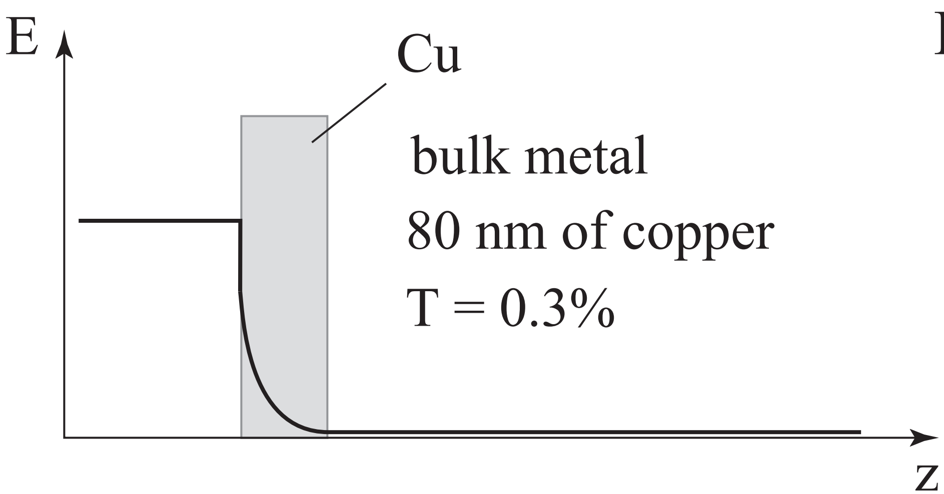
Also, metal-dielectric composites possess surface plasmon resonances, which can further enhance the NL response.

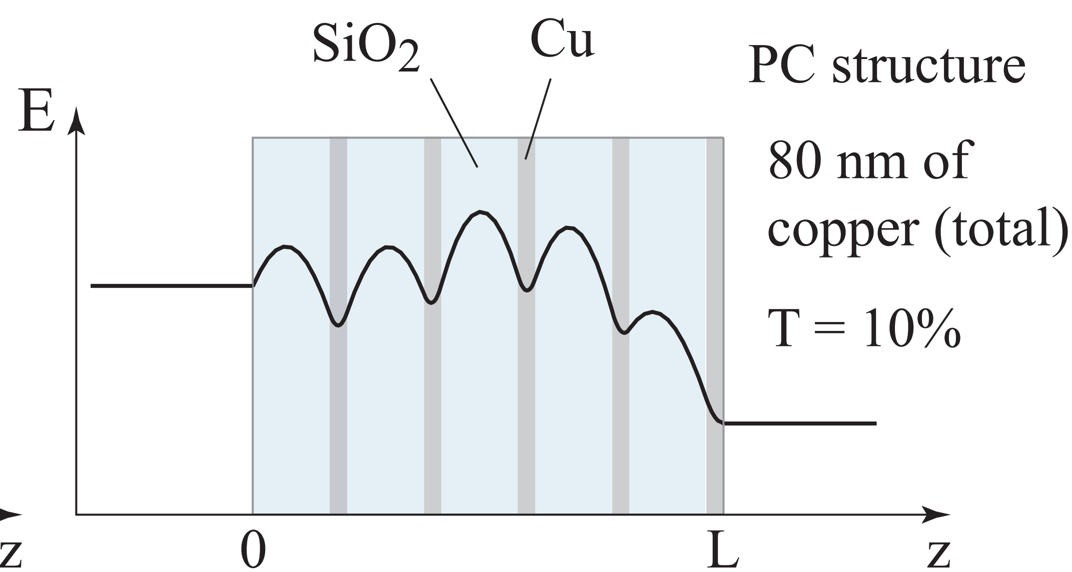
Comment 1: surface plasmons play no role in the work I am presenting today

Comment 2: I have worked on many of these approaches, see www.optics.rochester.edu/~boyd for details

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)

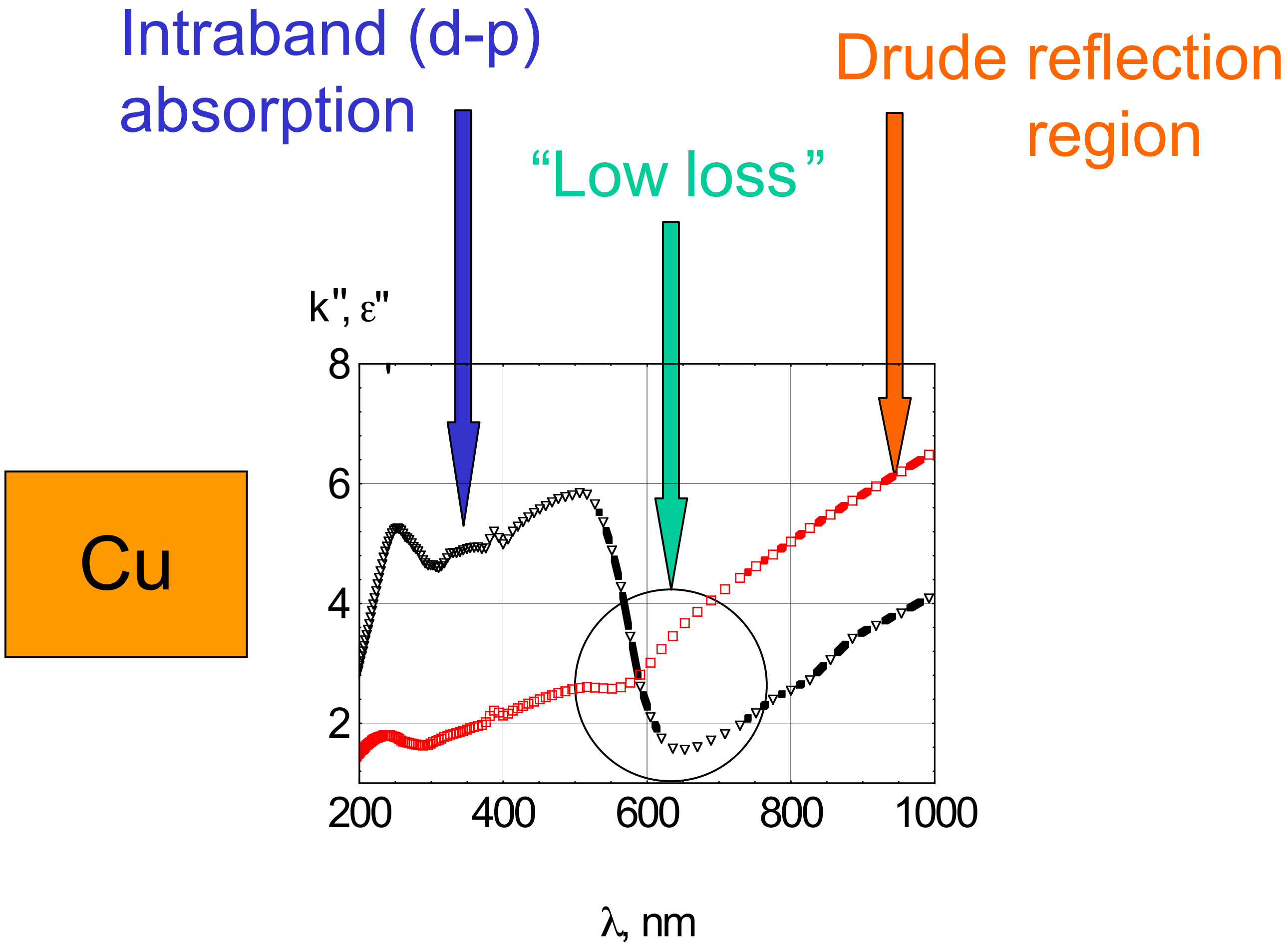




Greater than 10% enhancement of NLO response is predicted!

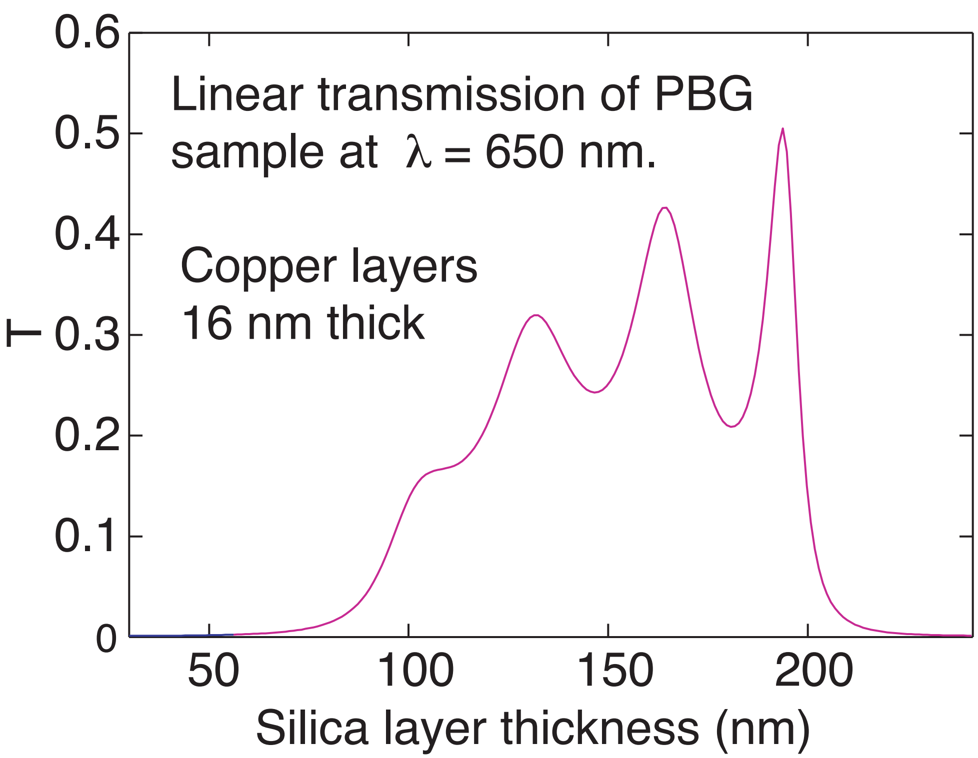
R.S. Bennink, Y.K. Yoon, R.W. Boyd, and J. E. Sipe, Opt. Lett. 24, 1416, 1999.

"Loss" mechanisms in copper

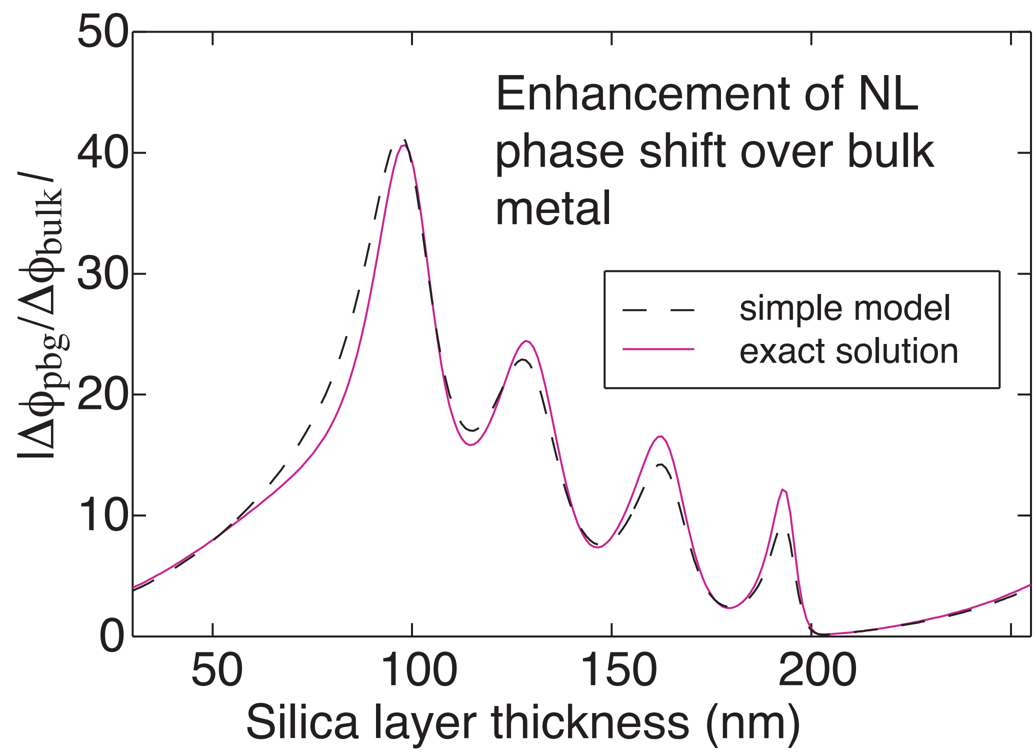


Accessing the Optical Nonlinearity of Metals with Metal-Dielectric Photonic Crystal Structures

- Metal-dielectric structures can have high transmission.



- And produce enhanced nonlinear phase shifts!

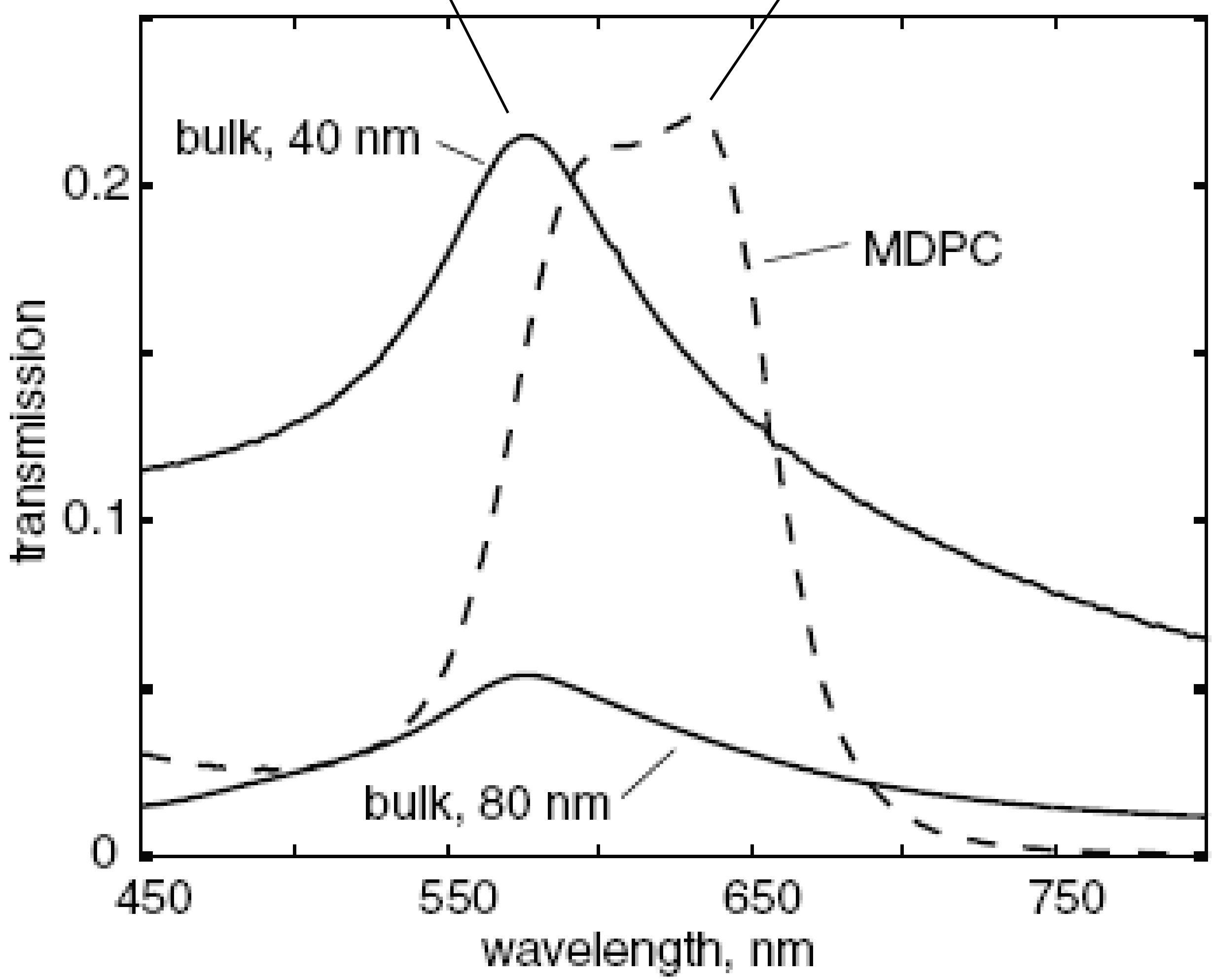


- Imaginary part of $\chi^{(3)}$ produces a nonlinear phase shift!
 (And the real part of produces nonlinear transmission!)

Linear Transmittance of Samples

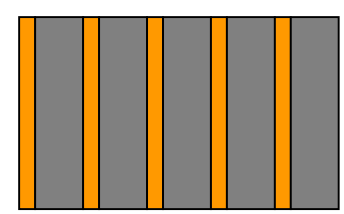
Material (interband) feature

Structural (M/D PC) feature



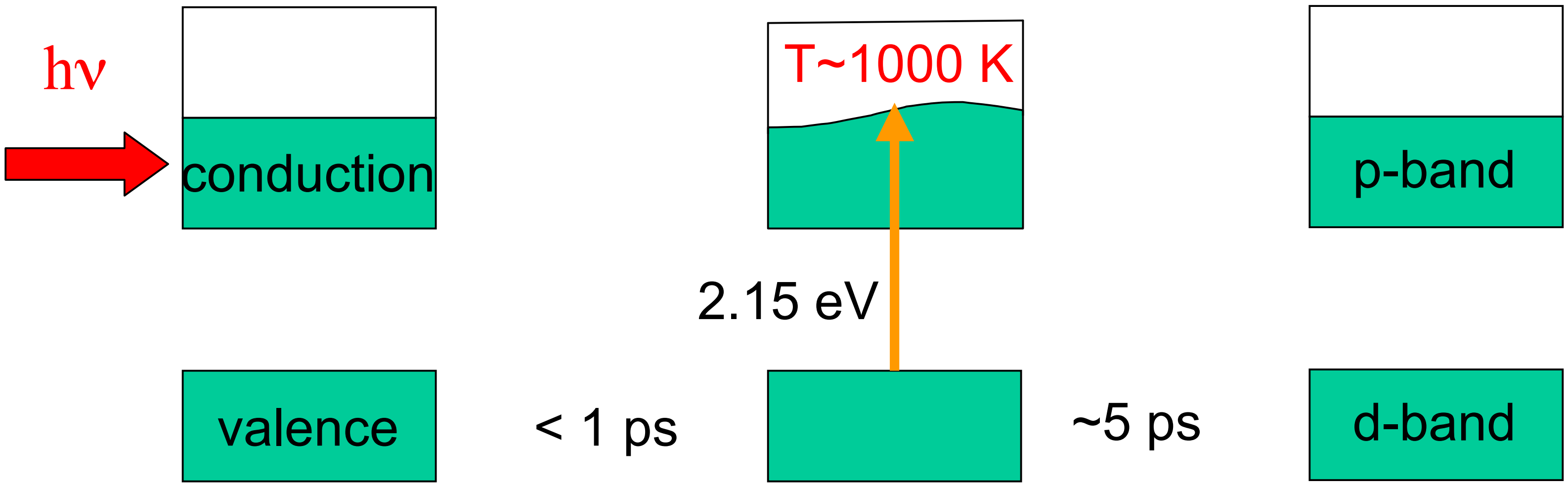
Cu: 40 nm film

M/D PC: Cu / silica



5x16/98 nm
(80 nm total Cu)

Mechanism of nonlinear response: "Fermi smearing"



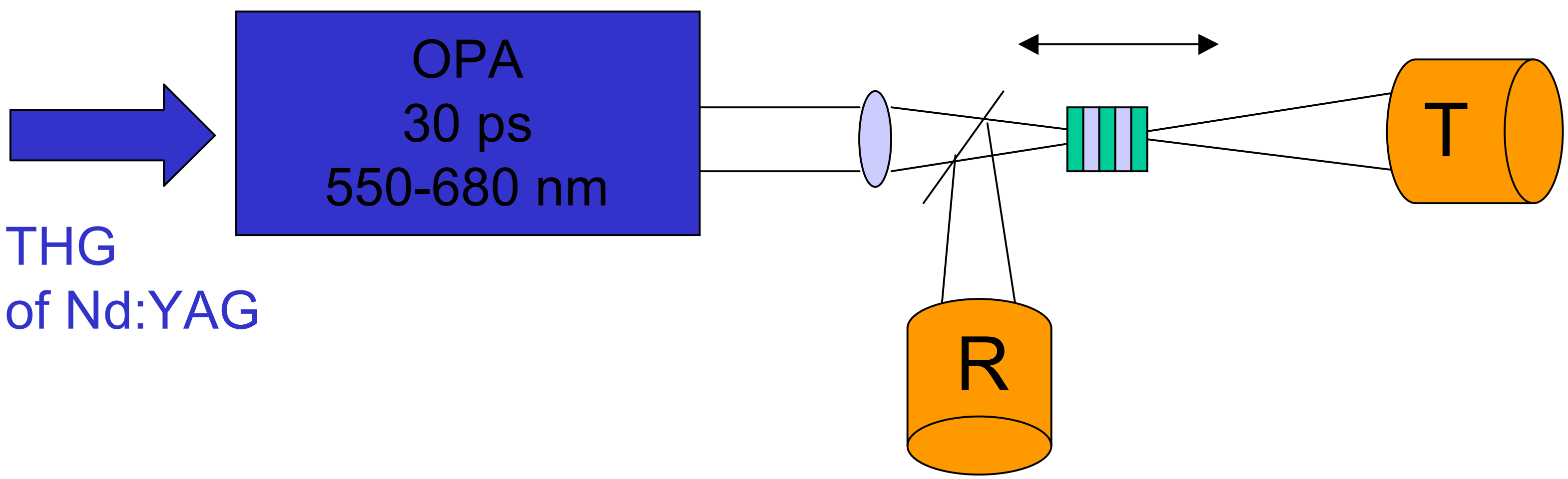
$$\Delta T \rightarrow \Delta\varepsilon(E_{IB}) \rightarrow \text{change in optical properties}$$

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)

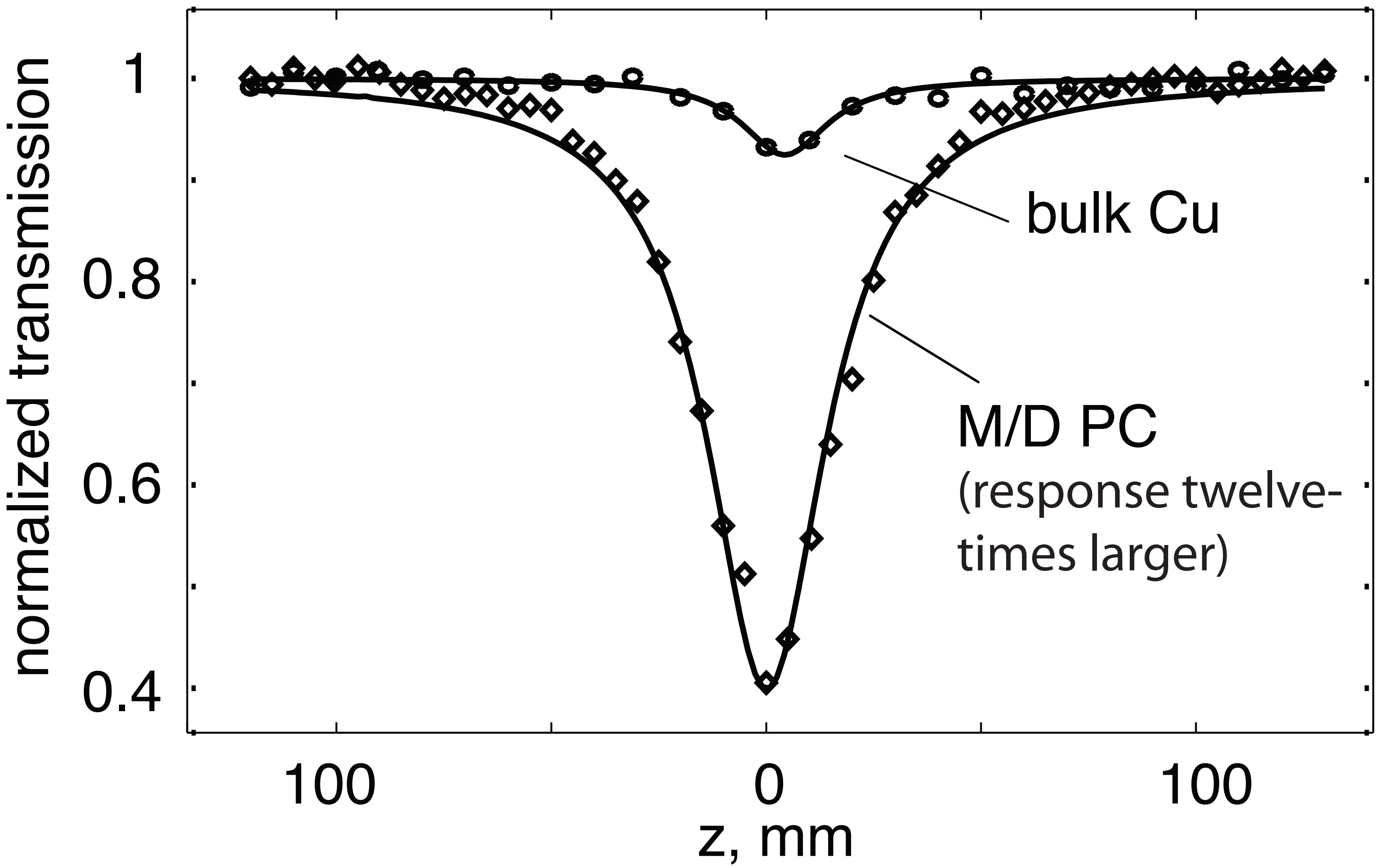
Reflection/Transmission Z-Scan



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



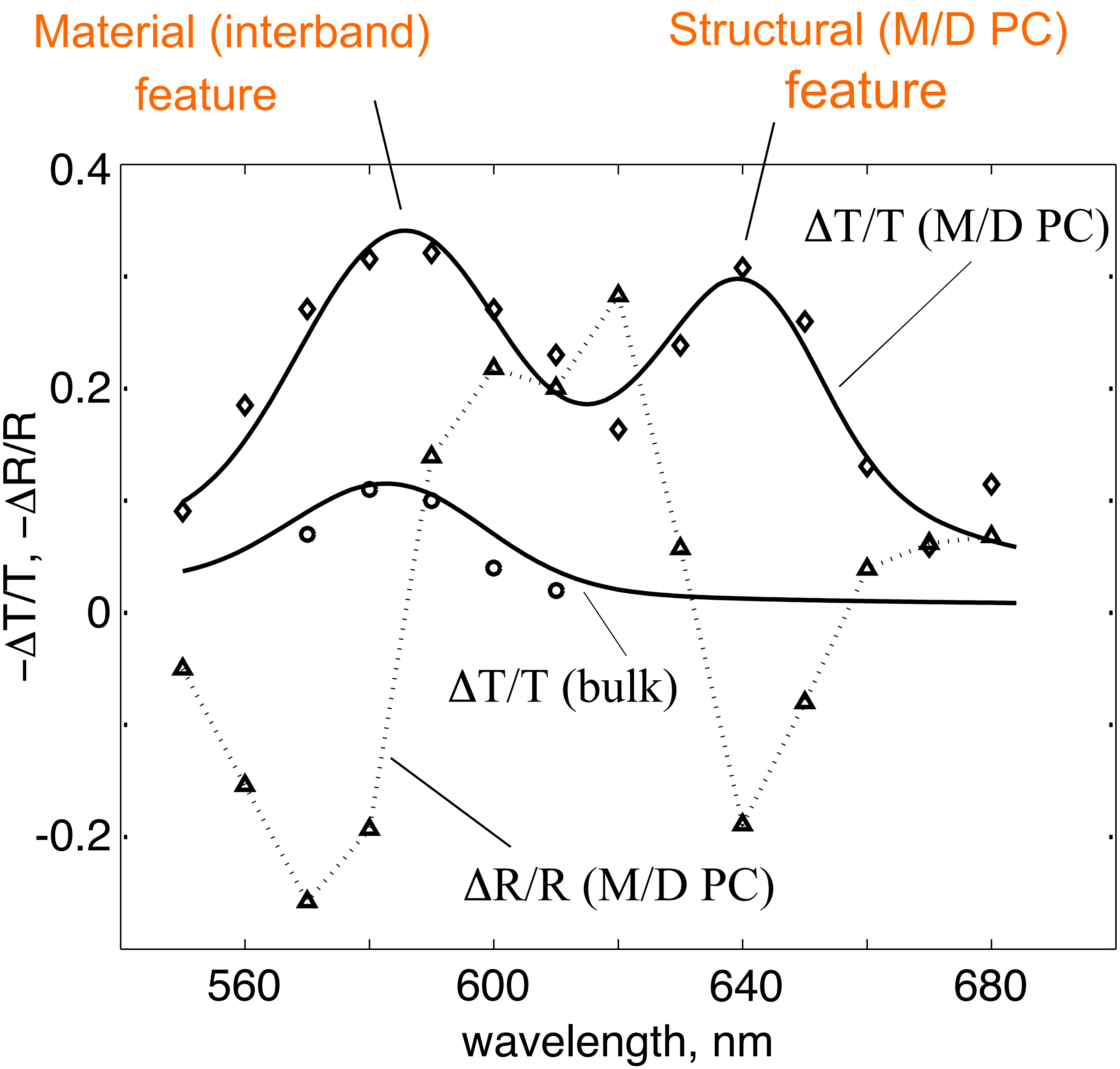
I = 500 MW/cm^2

λ = 650 nm

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

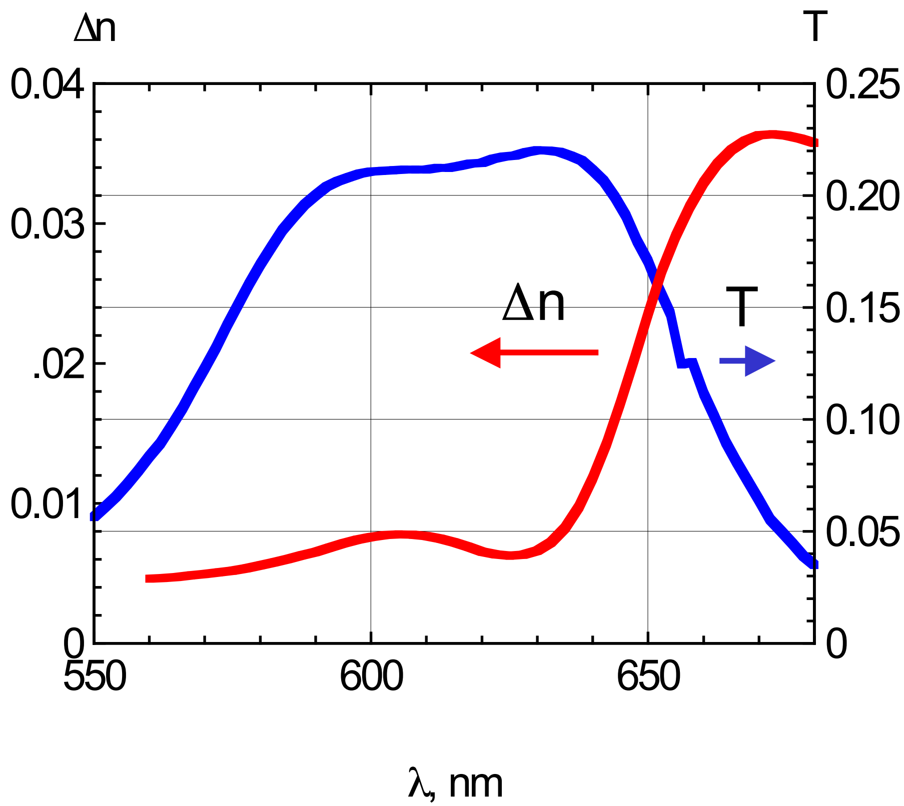
Lepeshkin, Schweinsberg, Piredda, Bennink, Boyd, Phys. Rev. Lett. 93 123902 (2004).

Nonlinear Transmission and Reflecance



Nonlinear phase shift in PC (numerical simulations)

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



Conclusions

- Stable, artificial, solid-state NLO material
- Enhanced transmission (10X)
- Enhanced nonlinear response in transmission (12X) over an extended spectral range (550-650 nm)
- Nonlinear phase shift resulting from $\Delta\varepsilon''$?

  Theory yes; experiment no.

  New design needed?