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!

“Loss” mechanisms in copper

- Intraband (d-p) absorption
- Drude reflection region

Plot showing k'' and ε'' against λ in nm.
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 $\chi^{(3)}$ produces nonlinear transmission!)

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**Linear transmission of PBG sample at $\lambda = 650$ nm.**

- Copper layers 16 nm thick

**Enhancement of NL phase shift over bulk metal**

- Simple model
- Exact solution
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

Reflection/Transmission Z-Scan

Pulse energy $\sim 1$ mJ
$I = 100$ MW/cm$^2$
We observe a large NL change in transmission
But there is no measurable NL phase shift for either sample

Nonlinear Transmission and Reflectance

Material (interband) feature

Structural (M/D PC) feature

$\Delta T/T (M/D PC)$

$\Delta R/R (M/D PC)$

$\Delta T/T (bulk)$

$-\Delta T/T, -\Delta R/R$

wavelength, nm

560  600  640  680
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?