Non-linear optics of metals at the interband absorption edge

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Introduction

$\chi^{(3)}$ - NLO properties of noble metals have been studied in:

- Nanoparticles
- Percolation films
- Thin films

Results:

$\chi^{(3)} >> \chi^{(3)}$ of silica
Sub-ps response time

Metals = losses!

Interaction length $L \sim$ skin depth

$\chi^{(3)}$ - mostly imaginary

Nonlinear response localized at the IB absorption edge
Artificial composite materials

Features:
Reduced loss (linear properties studied by Bloemer and Scalora [1])
Enhanced nonlinear response (theory by Bennink et al. [2])
Shifted peak of nonlinear response
Imaginary $\chi^{(3)} \rightarrow$ nonlinear phase shift
Increased damage threshold

Loss mechanisms in metals

Linear transmittance

Transmission

Cu
PBG

Cu: 40 nm film
PBG: 5x16/98 nm

Copper (80nm!) / silica
“Fermi smearing”

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

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

Reflection/Transmission Z-scan

Pulse energy $\sim 1\text{mJ}$
$I = 100\text{ MW/cm}^2$

\[ \frac{? R}{R},\quad \frac{\Delta T}{T} \rightarrow \Delta \varepsilon' + \Delta \varepsilon'' \rightarrow \chi_{\text{eff}}^{(3)} \]
Cubic susceptibility of pure Cu

\(\chi^{(3)}_{\text{eff}}, \, 10^{-8} \text{ esu}\)

Width of resonance \(\sim kT\)
Nonlinear response of PBG

\[ \frac{\text{Im}(\chi^{(3)}_{\text{PBG}})}{\text{Im}(\chi^{(3)}_{\text{Cu}})} \approx 12 \]

\[ \frac{\text{Re}(\chi^{(3)}_{\text{PBG}})}{\text{Re}(\chi^{(3)}_{\text{Cu}})} \approx 20 \]

Strong nonlinear features @ 650 nm!
Nonlinear phase shift in PBG

\[ \Delta \varepsilon = 0.1i \rightarrow ? f \]

Phase shift

Transmittance

\[ \Delta n \]

\[ \lambda, \text{nm} \]
Conclusions

• Stable, artificial, solid-state NLO material
• Enhanced transmission (10X)
• Enhanced nonlinear response (20X) over extended spectral range (550-650 nm)