Advanced Vitreous State – The Physical Properties of Glass

Active Optical Properties of Glass

Lecture 21: Nonlinear Optics in Glass-Applications

Denise Krol
Department of Applied Science
University of California, Davis
Davis, CA 95616
dmkrol@ucdavis.edu

Nonlinear optical susceptibilities

General formalism:

$$P(t) = \chi^{(1)}E(t) + \chi^{(2)}E(t)^2 + \chi^{(3)}E(t)^3 + \ldots$$
$$= P^{(1)}(t) + P^{(2)}(t) + P^{(3)}(t) + \ldots$$

E and P can be written as sum of frequency components:

$$E = \sum_j E(\omega_j)e^{-i\omega_j t} \qquad P = \sum_j P(\omega_j)e^{-i\omega_j t}$$

$$\chi^{(1)}(\omega) = \frac{P^{(1)}(\omega)}{\varepsilon_o E(\omega)} = \frac{Ne^2}{\varepsilon_o m}\frac{1}{\omega_0{}^2 - \omega^2 - i\omega\gamma}$$

output frequency

input frequencies, pos or neg

$$D(\omega_j) = (\omega_0^2 - \omega_j{}^2) - i\omega_j\gamma$$

$$\chi^2(\omega_p = \overbrace{\omega_m + \omega_n}) = \frac{P^{(2)}(\omega_p)}{E(\omega_m)E(\omega_n)} = \frac{Nae^3}{m^2 \cdot D(\omega_p)D(\omega_n)D(\omega_m)}$$

$$\chi^{(3)}(\omega_q = \omega_m + \omega_n + \omega_p) = \frac{Nbe^4}{m^3 \cdot D(\omega_q)D(\omega_m)D(\omega_n)D(\omega_p)}$$

Value of $\chi^{(n)}$ depends on frequencies

Nonlinear optics in glass

2nd-order nonlinearities

- In normal glasses $\chi^{(2)}=0$

3nd-order nonlinearities

- All materials, including glasses, have a $\chi^{(3)}$
- In glass there are only three independent $\chi^{(3)}$ tensor elements
- $\chi^{(3)}$ processes involve the interaction of 3 input waves to generate a polarization (4th wave) at a mixing frequency
  with 3 different input frequencies there are many possible output frequencies

$$\chi^{(3)}(3\omega = \omega + \omega + \omega) \neq \chi^{(3)}(\omega = \omega + \omega - \omega)$$

- Strength of generated signal depends on propagation length
  -optical fibers!
- Phase matching: $\Delta k = k_4 - k_3 - k_2 - k_1 = 0$

Units in nonlinear optics

Gaussian system of units

$$\vec{P}(t) = \underbrace{\chi^{(1)} \cdot \vec{E}(t)}_{\text{linear}} + \underbrace{\chi^{(2)} \cdot \vec{E}(t)^2 + \chi^{(3)} \cdot \vec{E}(t)^3 + \cdots}_{\text{nonlinear}}$$

MKS system

$$\vec{P}(t) = \varepsilon_0 \left[\chi^{(1)} \cdot \vec{E}(t) + \chi^{(2)} \cdot \vec{E}(t)^2 + \chi^{(3)} \cdot \vec{E}(t)^3 + \cdots \right]$$

ε_0 = permittivity of free space = 8.85 x 10^{-12} F/m

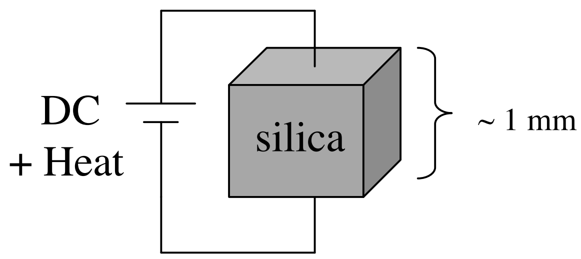
	MKS system	Gaussian system
Electric Field, E	V/m	statvolt/cm
Polarization, P	C/m^2	statvolt/cm
Intensity, I	$I = 2n\left(\frac{\varepsilon_0}{\mu_0}\right)^{1/2} \lvert E \rvert^2$	$I = \frac{nc}{2\pi} \lvert E \rvert^2$
Intensity, I	W/m^2	erg/cm^2-sec
$\chi^{(2)}$	m/V	cm/statvolt, esu
	$\chi^{(2)}$ (MKS) = 4.189 x 10^{-4} $\chi^{(2)}$ (Gaussian)	
$\chi^{(3)}$	m^2/V^2	$(cm/statvolt)^2$, esu
	$\chi^{(3)}$ (MKS) = 1.40 x 10^{-8} $\chi^{(3)}$ (Gaussian)	

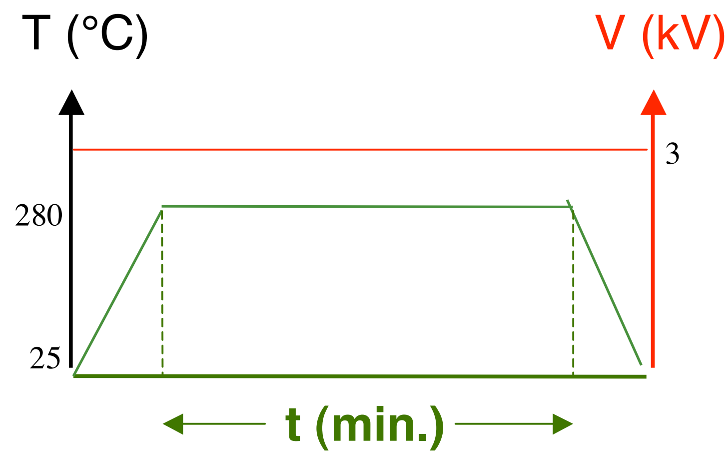
$\chi^{(2)}$ can be induced in glass by thermal poling

Second order optical nonlinearity ($\chi^{(2)}$) = 0 in glasses because glasses are isotropic

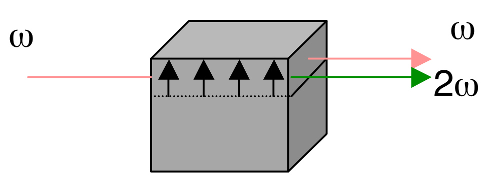
To induce a $\chi^{(2)}$ in glasses → Thermal poling technique

Thermal poling experiment



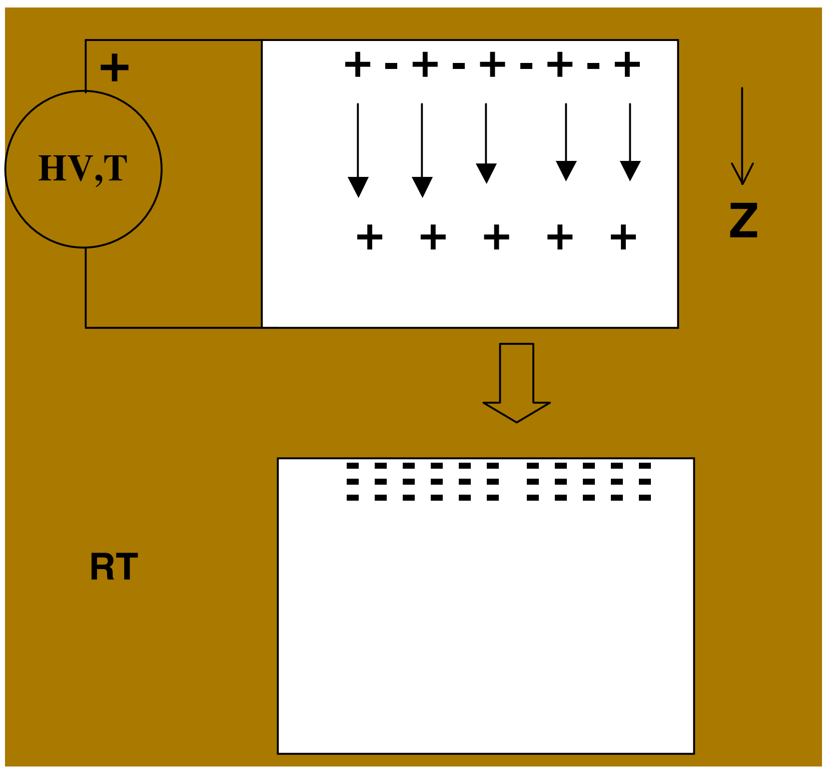


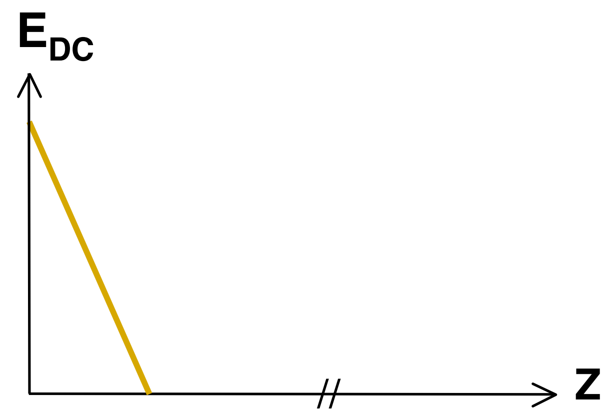
SHG process

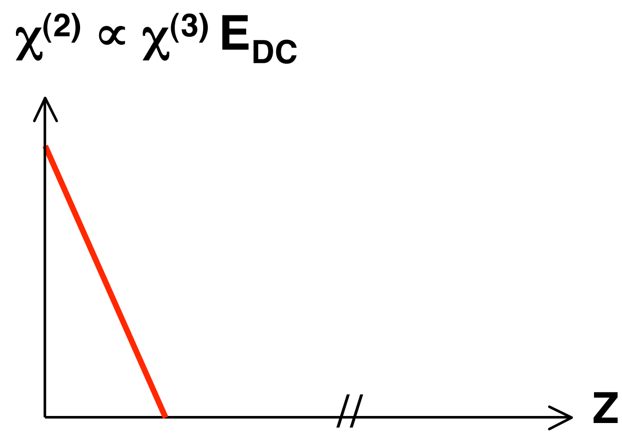


The induced $\chi^{(2)}$ can be examined via second harmonic generation (SHG)

Thermal poling-proposed mechanism



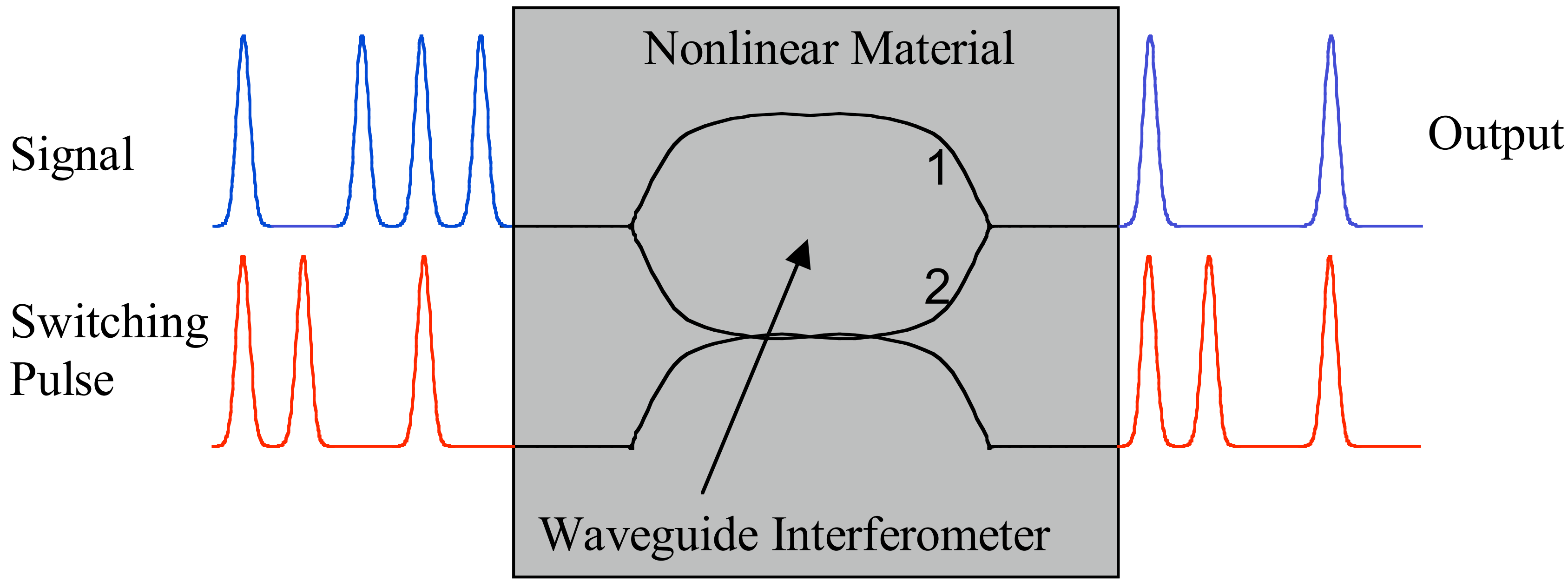




$\chi^{(3)}$ phenomena and applications in glass

Effect		Applications
Nonlinear index	$n = n_0 + n_2 I$ $n_2 \sim \chi^{(3)}(\omega = \omega + \omega - \omega)$	**Optical switching** **Supercontinuum generation**
Stimulated Raman scattering		**Raman amplifiers and lasers**
Nonlinear photoinduced changes		**Fs laser structuring**

Nonlinear optical switch



Without switching pulse: waves in leg 1 and 2 interfere destructively, no output

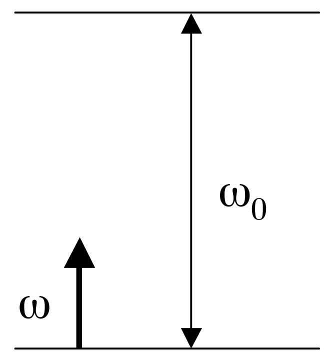
With switching puse: due to the nonlinear interaction, the switching pulse causes a phase shift in the part of the signal pulse propagating in leg 2. As a result waves in 1 and 2 interfere constructively, output

From P.Thielen, PhD Dissertation, UC Davis, 2004

Material dependence of n_2

Classical anharmonic electron oscillator, far from resonance:

$$\chi^{(3)}(\omega = \omega + \omega - \omega) \approx \frac{e^4}{m^3 \omega_0{}^6 d^5}$$



Bond polarizability model by M. Lines:

Long wavelength limit:

$$n_2(0) = \frac{3.4(n_0{}^2 + 2)^3 (n_0{}^2 - 1) d^2}{n_0{}^2 E_S{}^2} \cdot 10^{-20}$$

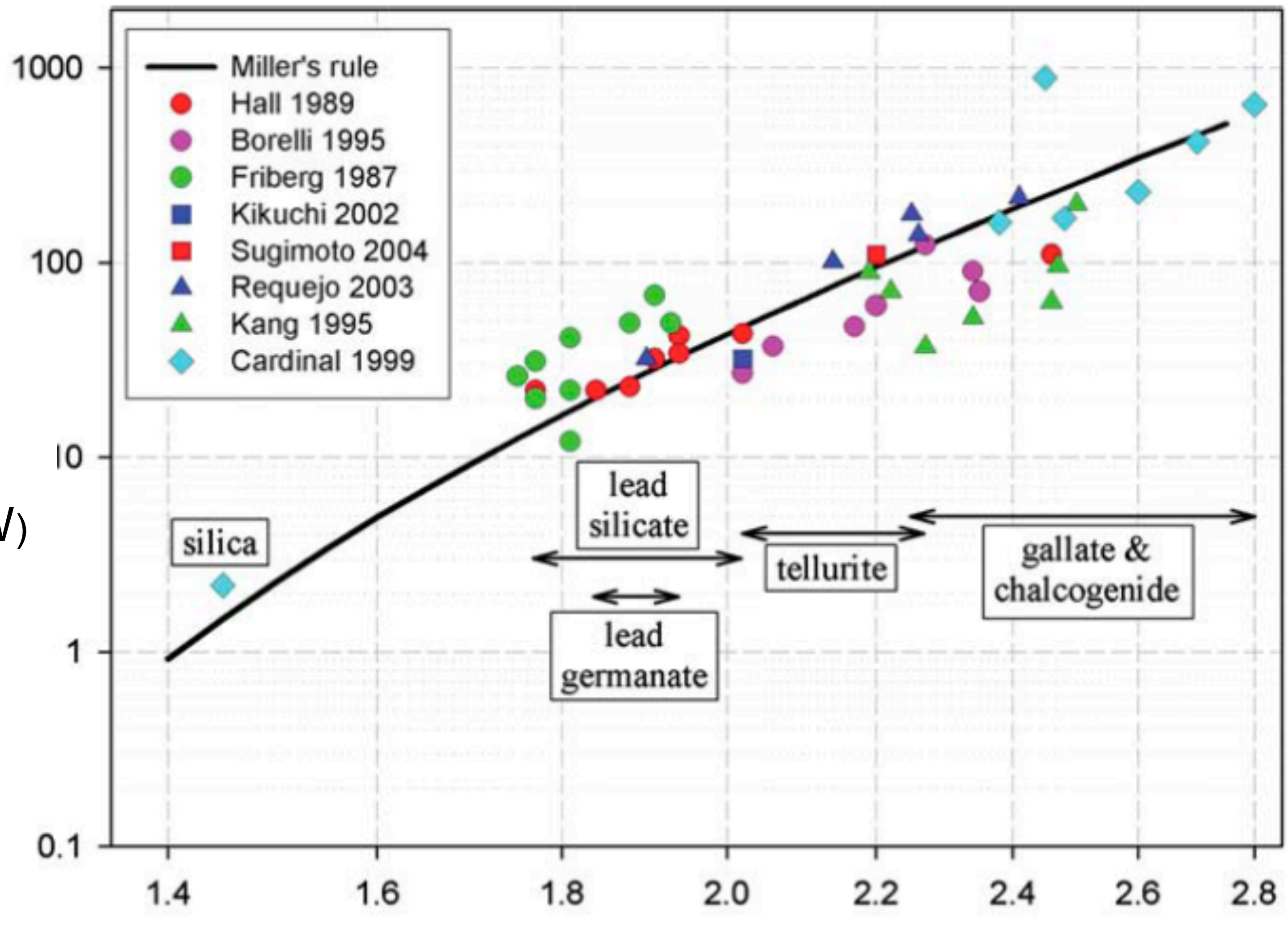
E_s is Sellmeier gap

Frequency dependence

$$n_2(\omega) = n_2(0) \cdot \left[1 - \left(\frac{\hbar\omega}{E_S} \right)^2 \right]^{-3.5}$$

Material dependence of nonlinear index

n_2
(10^{-16}cm^2/W)

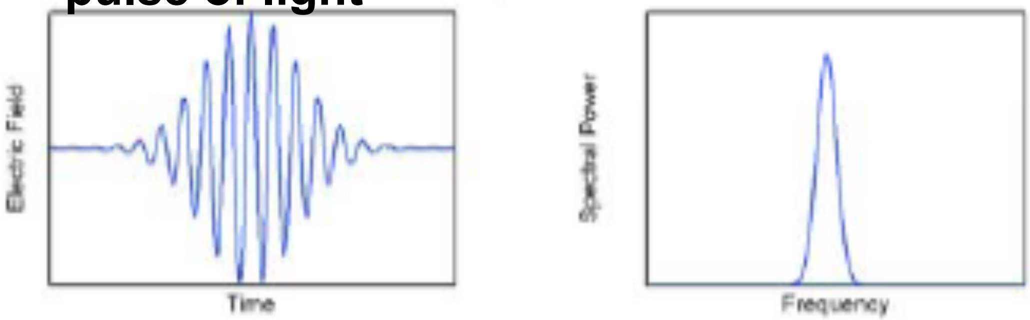


T. Monro et al, Annu. Rev. Mater. Res. 2006. 36:467

n_0

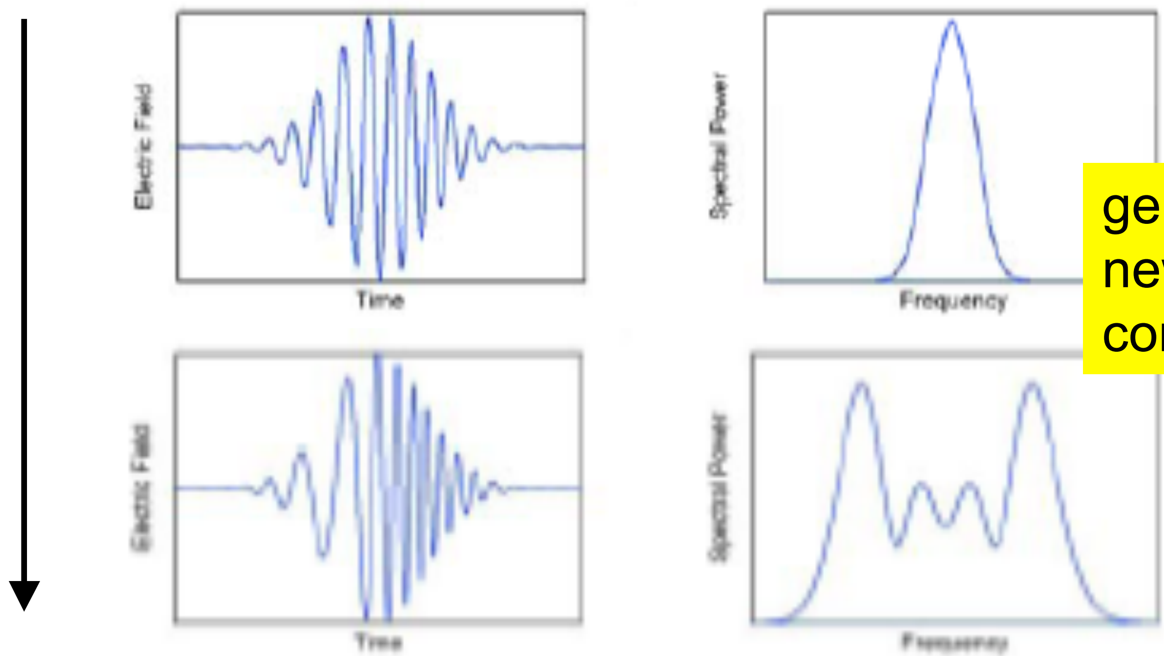
self phase modulation

pulse of light



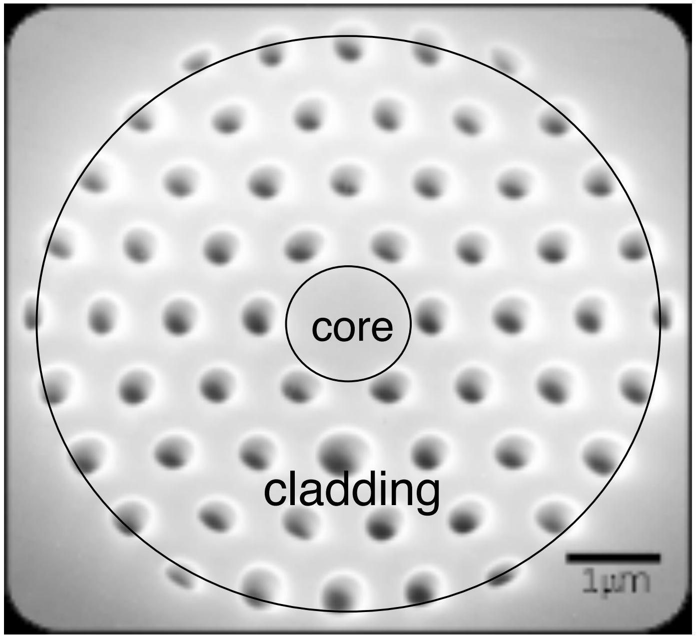
instantaneous frequency $$\varpi(t) = \omega_0 - \frac{\omega_0 n_2 L}{c} \frac{dI(t)}{dt}$$

distance through fiber

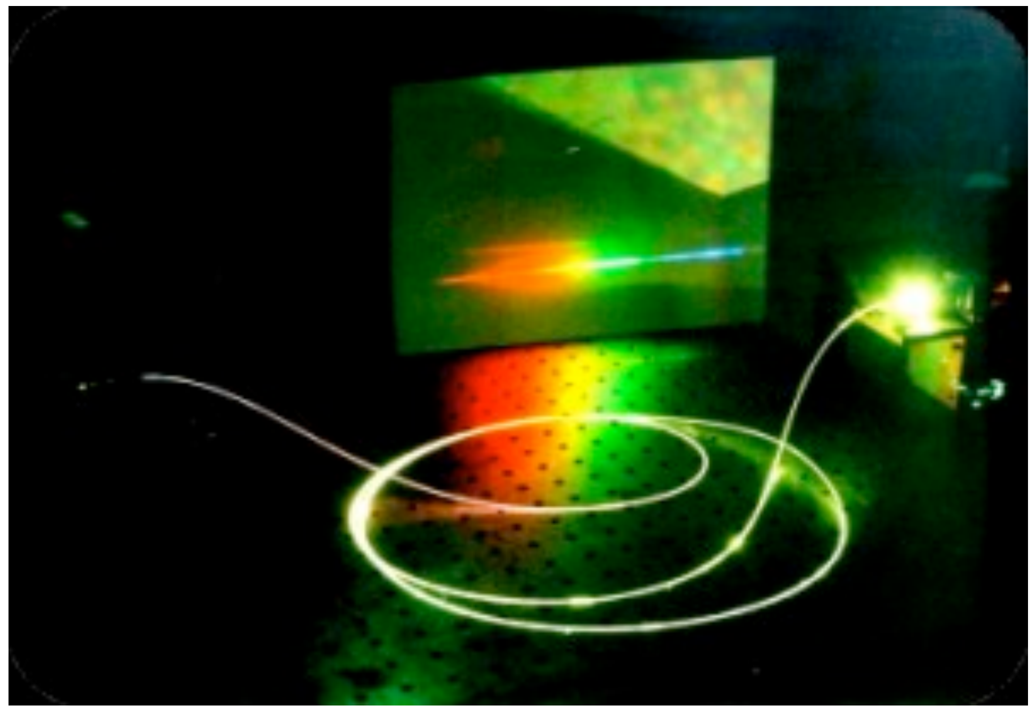


generation of new frequency components

Supercontinuum generation in microstructured fibers



propagation of pulsed (100fs) Ti-sapphire laser light(800 nm) results in supercontinuum generation : 400-1600 nm



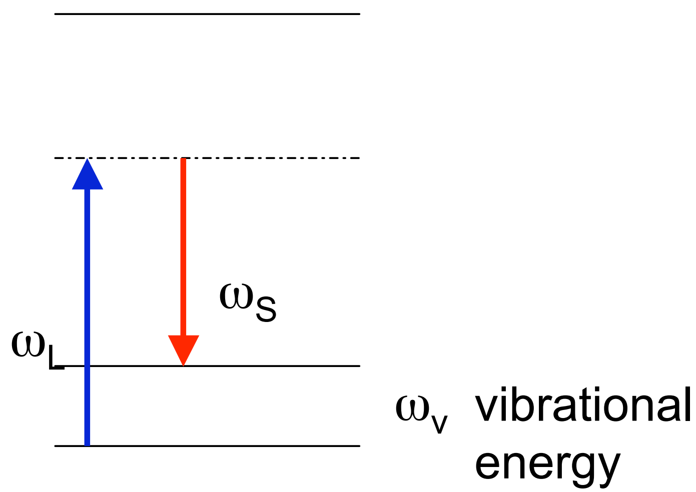
guidance properties determined by size and pattern of holes

unusual dispersion
high nonlinearity

From Philip Russell et al. Source:www.bath.ac.uk/physics/groups/opto/rse/holeyfibres.html

Raman gain

Stokes Raman scattering



At high laser intensities: stimulated Stokes Raman scattering

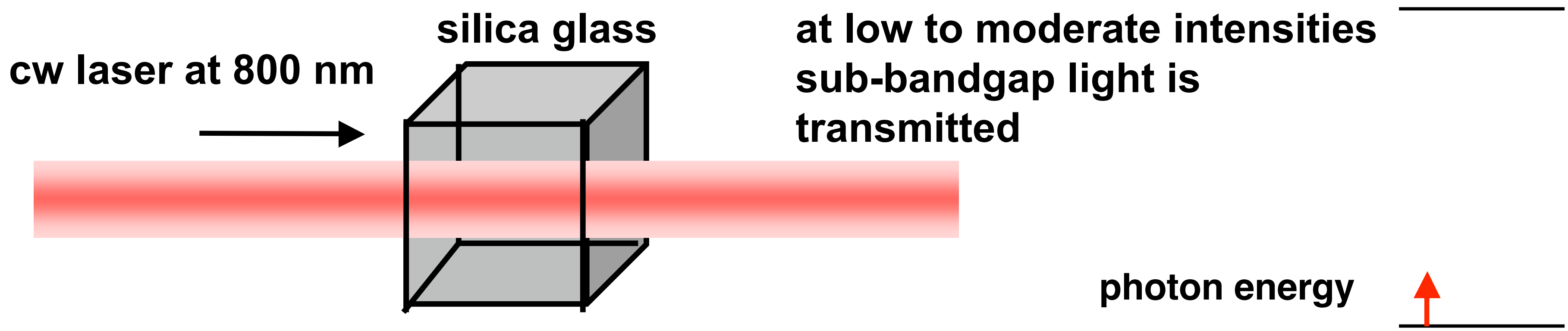
$$P^{NL}(\omega_S, z) = 6\chi_R(\omega_S)\left|A_L\right|^2 A_S e^{ik_S z}$$

$$\chi_R(\omega_S) = \frac{N}{6m}\left(\frac{\partial\alpha}{\partial q}\right)_0^2 \cdot \frac{1}{\omega_v^2 - (\omega_L - \omega_S)^2 + 2i\gamma(\omega_L - \omega_S)}$$

$\chi^{(3)}$ phenomena and applications in glass

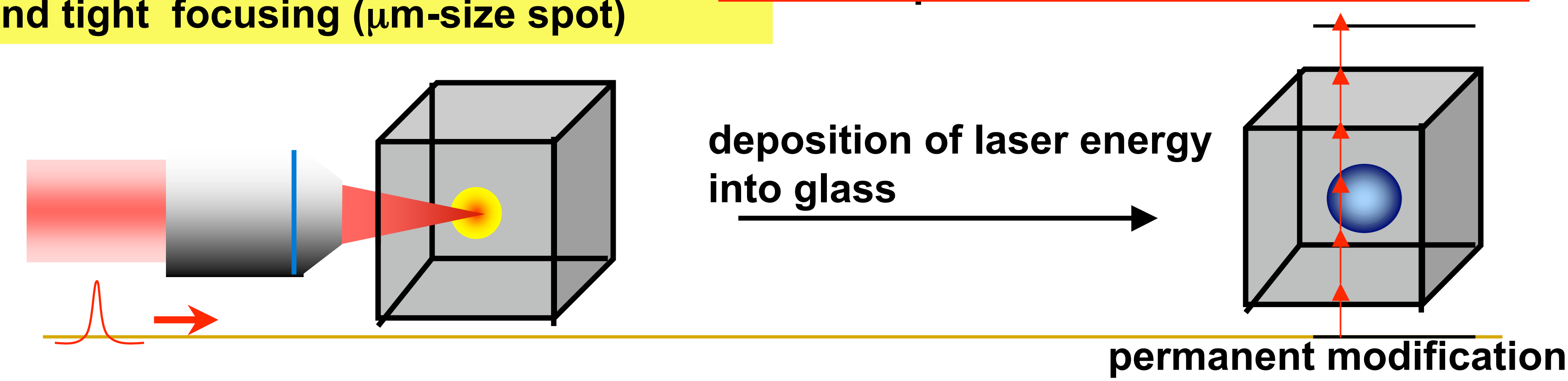
Effect	Applications
Nonlinear index	**Optical switching** **Supercontinuum generation**
Stimulated Raman scattering	**Raman amplifiers and lasers**
Nonlinear photoinduced changes	**Fs laser structuring**

Interaction of glass with sub-bandgap, focused, fs laser pulses

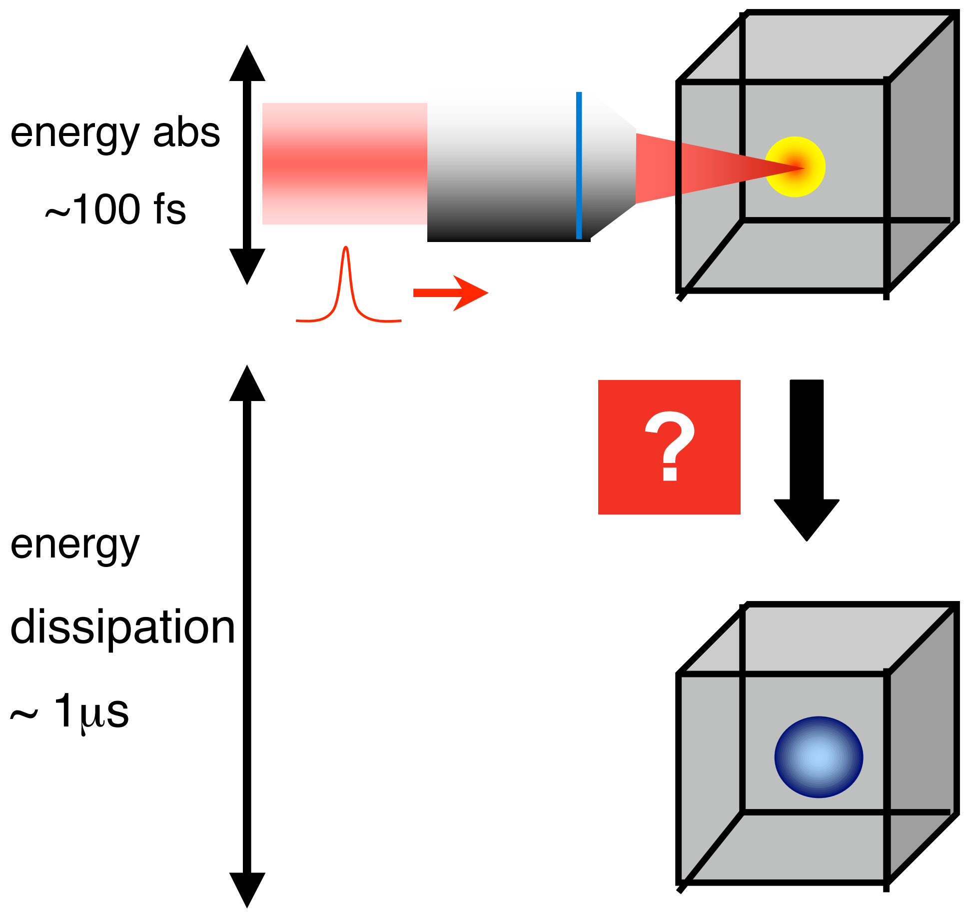


Light-matter Interaction is localized in time and space -> 3-D control of modification

ultrashort (100fs) pulses and tight focusing (μm-size spot)



Femtosecond laser modification in glass



1) Multiphoton absorption
2) Avalanche photoionization
3) Plasma formation

4) Proposed mechanism:
-Shockwave propagation (microexplosion)
-Fast heating and cooling

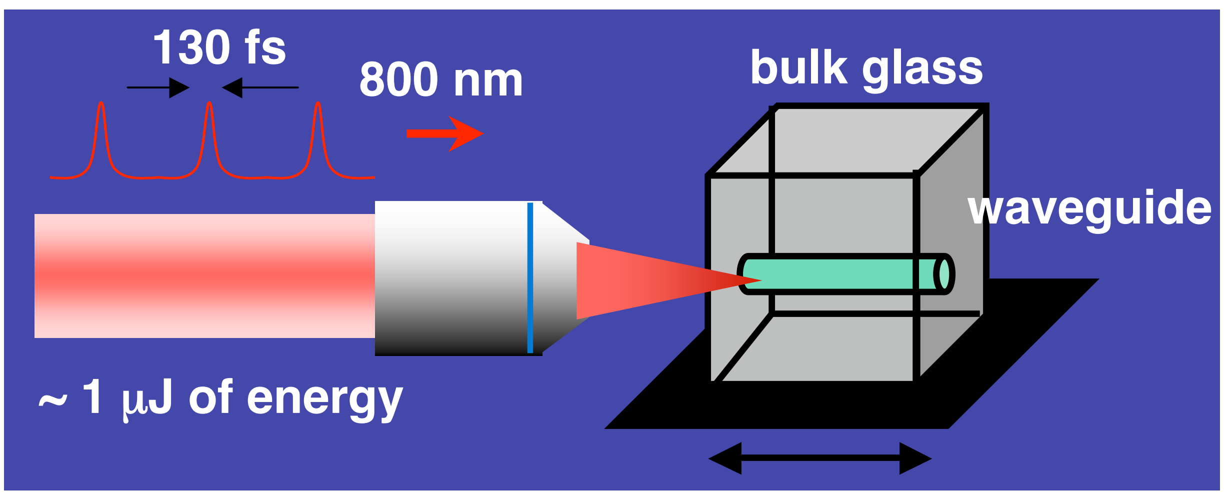
5) Modified spot

How does the material change on an atomic scale?

Schaffer et al, MRS Bull 31, 620 (2006)

Femtosecond laser pulses can modify various glass properties

Properties:	**Applications:**
➢ **Refractive index**	**photonic devices**
➢ **Absorption**	
➢ **Composition (phase separation)**	**lab-on-chip**
➢ **Valence state (Sm3+ -> Sm2+)**	**data storage**
➢ **Crystal nucleation (Ag and Au colloids in glass)**	**optical switching**



Davis *et. al*, Opt. Lett., 21, 1729 (1996)

Some references

NLO Books:

N. Bloembergen, Nonlinear Optics

R.W. Boyd, Nonlinear Optics

NLO in Glass Reviews

E. M. Vogel, M.J. Weber, D. M. Krol, “Nonlinear optical phenomena in glass”, Phys. Chem. Glasses 32, 231 (1991).

K. Tanaka, “Optical nonlinearity in photonic glasses”, J. Materials Science: 16, 633 (2005)

Fs laser structuring of glass

“Ultrafast lasers in materials research”, Special issue, MRS Bulletin August 2006