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All-Paid US-Japan Winter School
on
New Functionalities in Glass

TOHOKU UNIVERSITY

Photonic Glass

Controlling Light with Nonlinear Optical Glasses and Plasmonic Glasses

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Outline

1) Background & Motivation
2) 2nd-order optical nonlinearity in glass
   -Controlling light with change of refractive index
3) Toward real application of electro-optic glass devices;
   -"UV-poling" and Permanent $\chi^{(2)}$
4) Recent topics of our research works
   -New EO glasses and fiber-type devices
   -"Plasmonic Glass", light localization / propagation

Motivation

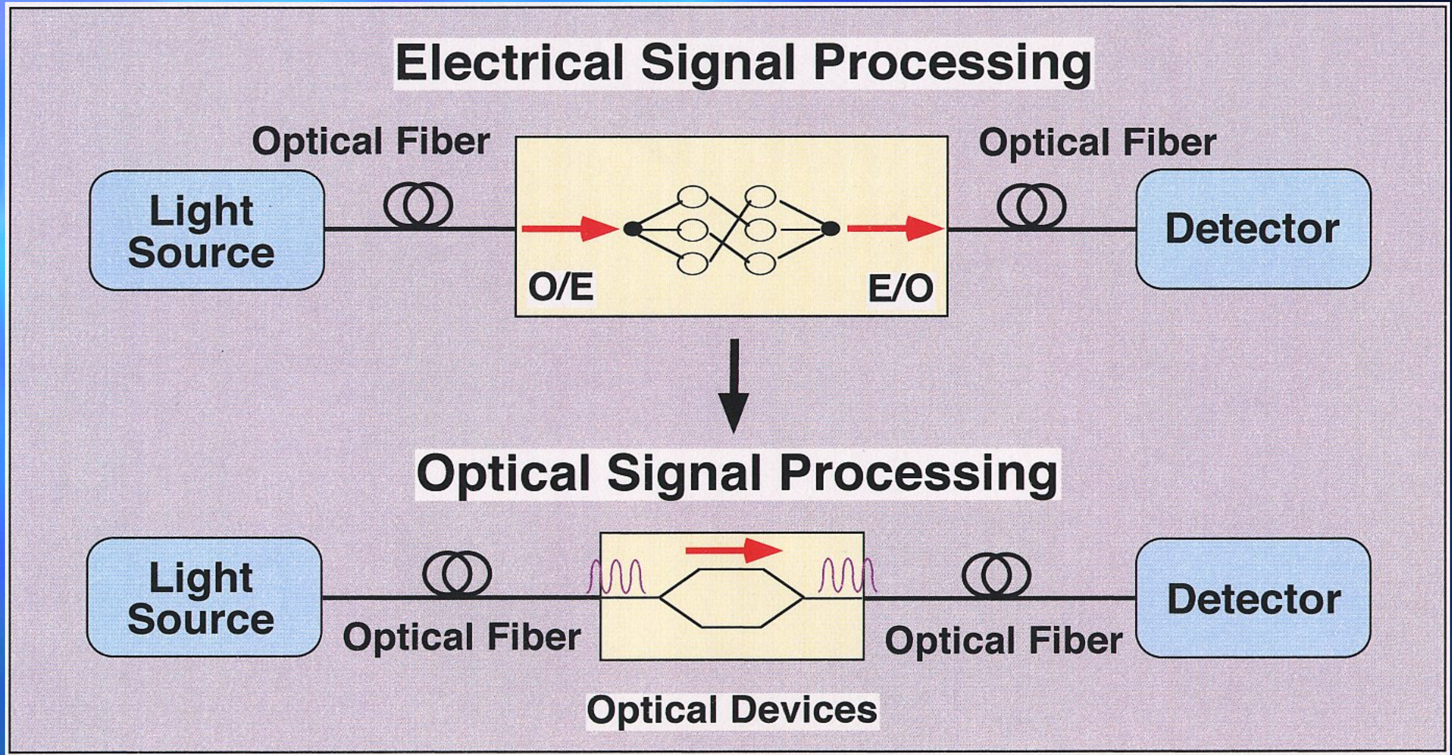
Novel nonlinear "glass materials" for photonic applications

Glass ⇒ key material

- High and wide range of transparency
- Good connectivity to glass fiber
- High environmental durability
- Easy shaping to fiber and films

... but not applicable for signal processing such as optical switching and modulation etc.

Advanced Photonic Communication



Functional Photonic Devices/Components with excellent connectivity to the fiber

E/O-Switch, Modulator, Converter, etc drived by *Second-Order Optical Nonlinearity*

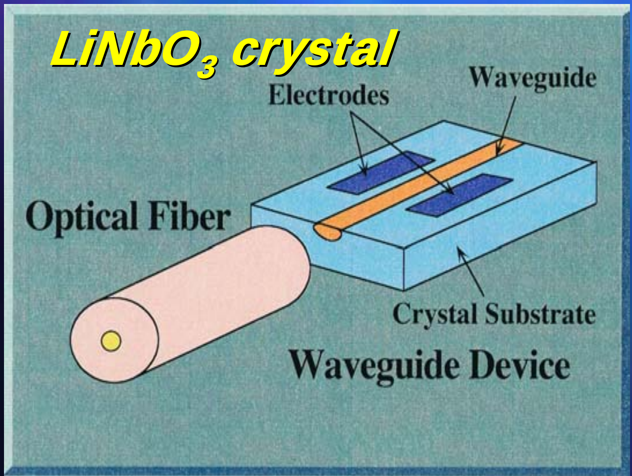
Second-Order Optical Nonlinearity in Glass

2^{nd}-order optical nonlinearity

$$P = \varepsilon_0 (\chi^{(1)} E + \chi^{(2)} EE + \chi^{(3)} EEE + \cdots)$$

P : polarization, ε_0: dielectric constant, E: electric field of light

2^{nd}-order nonlinearity is NOT allowed in glasses with inversion-symmetry



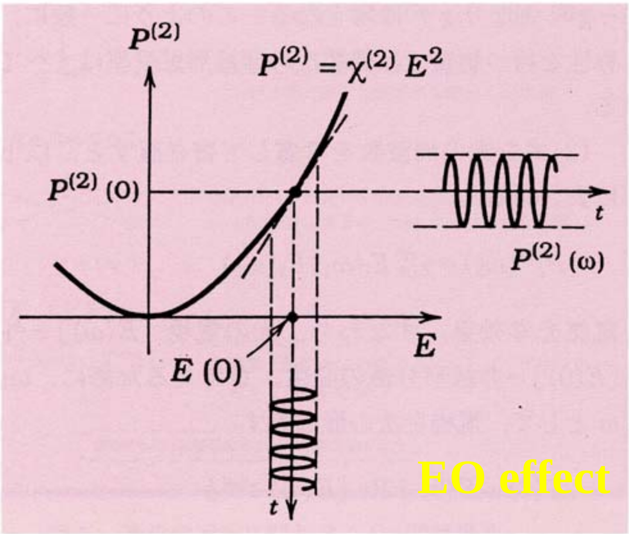
Permanent connection to *glass-fibers*

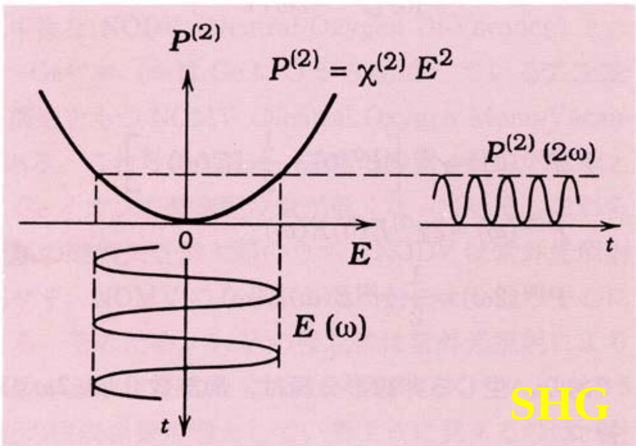
glass-crystal connection

Photonic Glass

Glass with 2^{nd}-order nonlinearity

Alternative Description of Δn





EO effect (Pockels effect)

Electric field of angular frequency : E(ω)
Applied electric field : E(0)
Nonlinear susceptibility : $\chi^{(2)}$

If E(0) > E(ω), at E=E(0)

$$P^{(2)} = \Delta\chi E(\omega)$$

where $\Delta\chi = 2\chi^{(2)}E(0)$ represents an increase in the susceptibility proportional to the electric field E(0).

The corresponding incremental change of the refractive index is obtained by the relation $n^2 = 1 + \chi$, to obtain $2n\Delta n = \Delta\chi$, from which

$$\Delta n = (\chi^{(2)}/n)E(0)$$

$\Delta n = -rn^3E/2$ is defined in the Pockels effect, thus, EO coefficient r is described by

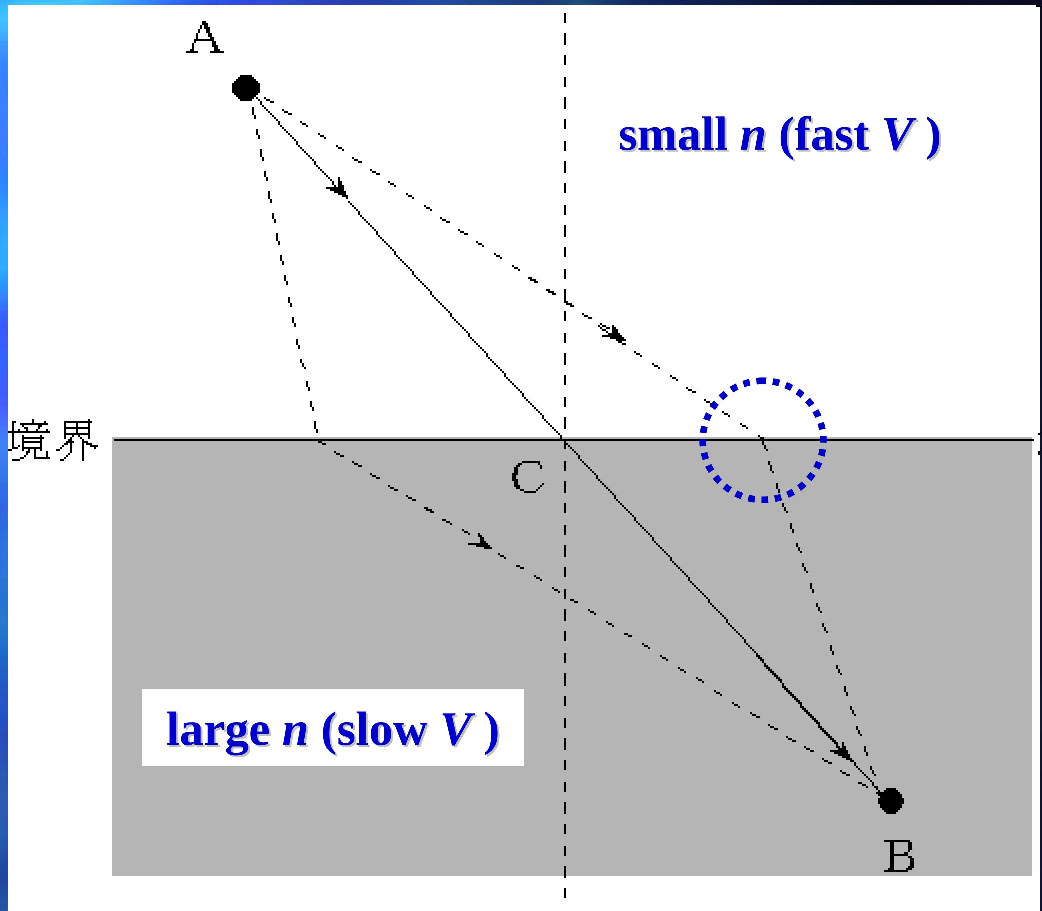
$$r = -2\chi^{(2)}/n^4$$

Fermat's Principle: Boundary Refraction

Speed of light:

$$V = C_0 / n$$

velocity in the medium: V
free space velocity: C_0
refractive index: n



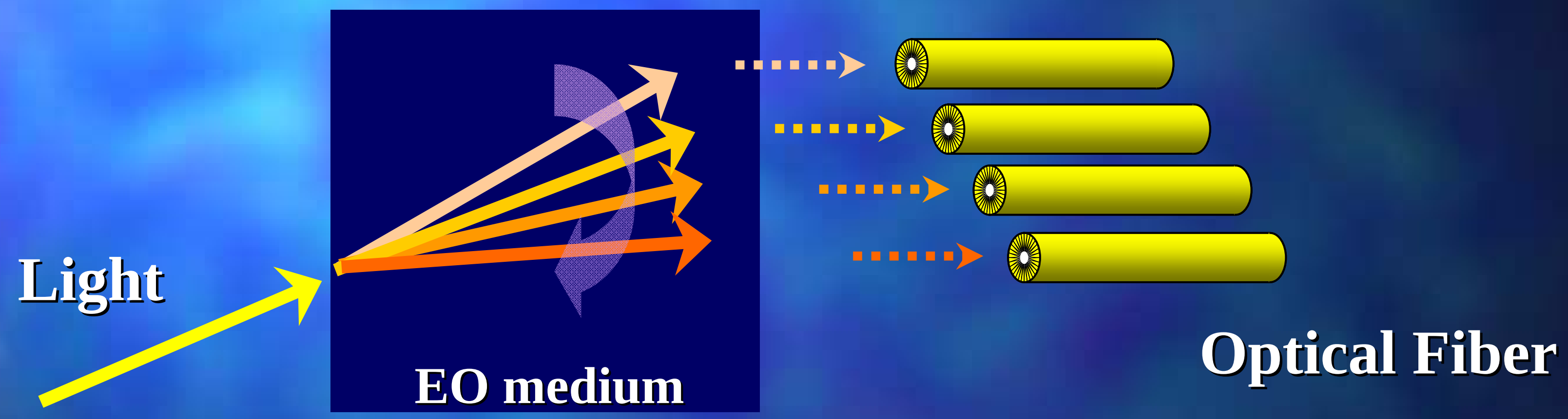
large $n \rightarrow V$: slow
small $n \rightarrow V$: fast

Light rays travel along the path of least time by refraction in this case (Snell' Law: $\sin\theta / \sin\phi = n_L / n_S$)

Controlling Light with EO Effect

Change of refractive index (Δn)

Angle of refraction changed by E_{appl}

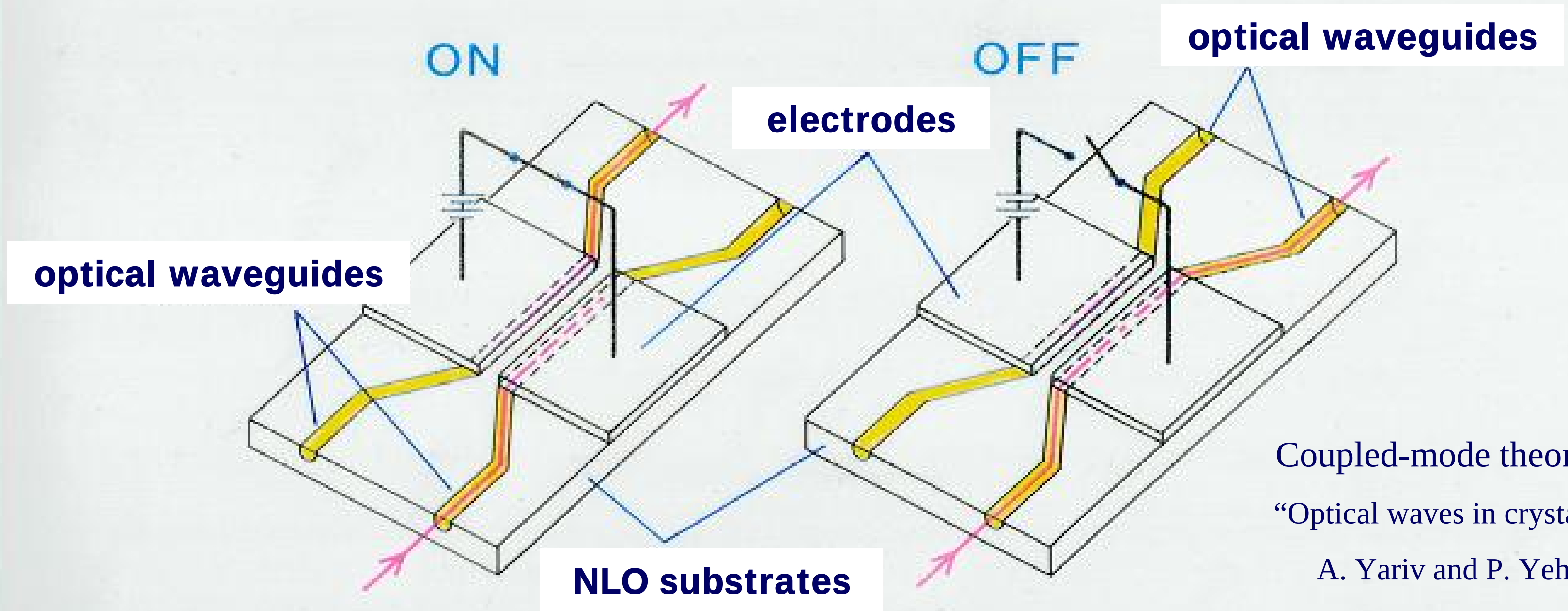


Cotrolling light with EO devices through 2nd order optical nonlinearity

Electro-Optic Devices

Directional Coupler

2x2 Optical Switch



Coupled-mode theory :

"Optical waves in crystals"

A. Yariv and P. Yeh

Advantages of "*Photonic Glass*"

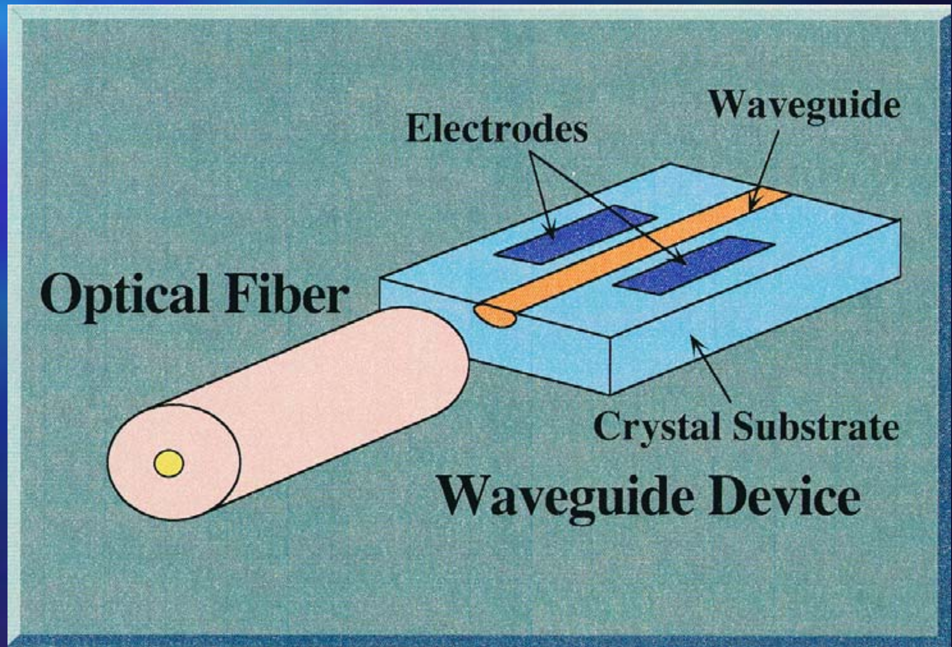
Advantages & Disadvantages

	Glasses	Inorganic materials	Organic materials
2nd-Order NL	X	○	◎
Optical Loss	◎	○	△
Transparent Range	◎	△	△
Material Design	○	X	◎
Connection	◎	X	○
Shaping	◎	△	△
Durability	◎	△	△

Photonic glass* is the best solution for glass-fiber networks.

***Second-Order Optical Nonlinearity**

- Long-term stability
- Low excess loss
- Easy to connect



How to induce $\chi^{(2)}$?

2^{nd}-order nonlinearity induced in glass

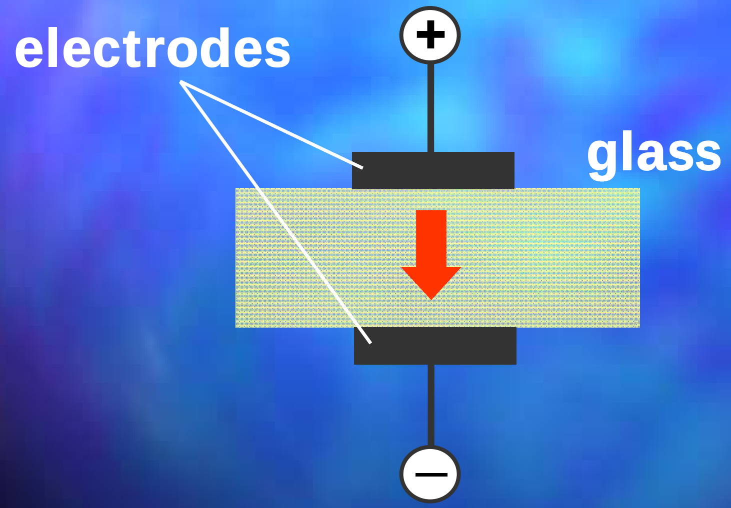
1. Poling with UV/heating
2. Crystallization

	Poling	*Crystallization*
$\chi^{(2)}$ value	**~10 pm/V**	~1 pm/V
patterning	**Yes (UV)**	No
stability (no decay)	**No**	**Yes**

$LiNbO_3$: ~28 pm/V

Poling in Glass/Fiber

Breaking of inversion symmetry in glass



Poling in glass…

Applied electric field

- **-At elevated temperature**
- **-With UV-laser irradiation**

Field-Induced Microstructuring in Glass Materials

UV-Poling in Glass / Fiber

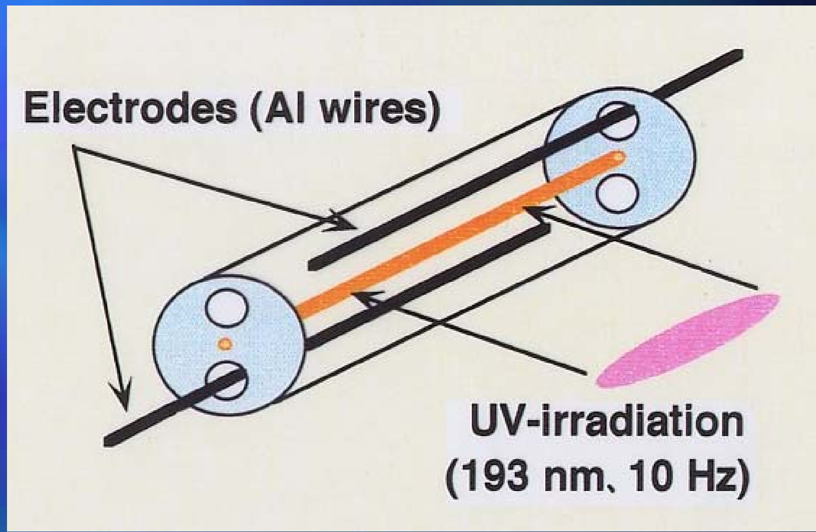
The Optical Fibre Technology Centre (OFTC)
University of Sydney, Australia

Thermal Poling

Poling Techniques	Composition and Form	$\chi^{(2)}$ or r (pm/V)	References
Photoinduced Poling	Ge-P-doped SiO_2 Fiber	$\chi^{(2)} \sim 10^{-4}$	Österberg (1986)
Room-Temperature Poling	Ge-P-doped SiO_2 Fiber	$r \sim 10^{-3}$	Li (1989)
Poling at Elevated Temperature (Thermal Poling)	-Fused Silica	$\chi^{(2)} \sim 1$	Myers et al. (1991)
	-Ge-doped SiO_2 fiber	$\chi^{(2)} \sim 0.2$	Kazansky (1991)
	-Ge-doped SiO_2 waveguide	$\chi^{(2)} \sim 0.5$	Liu (1994)

$\chi^{(2)}$ **was limited by <1pm / V**

UV-Poling in Ge:SiO_2 Fiber

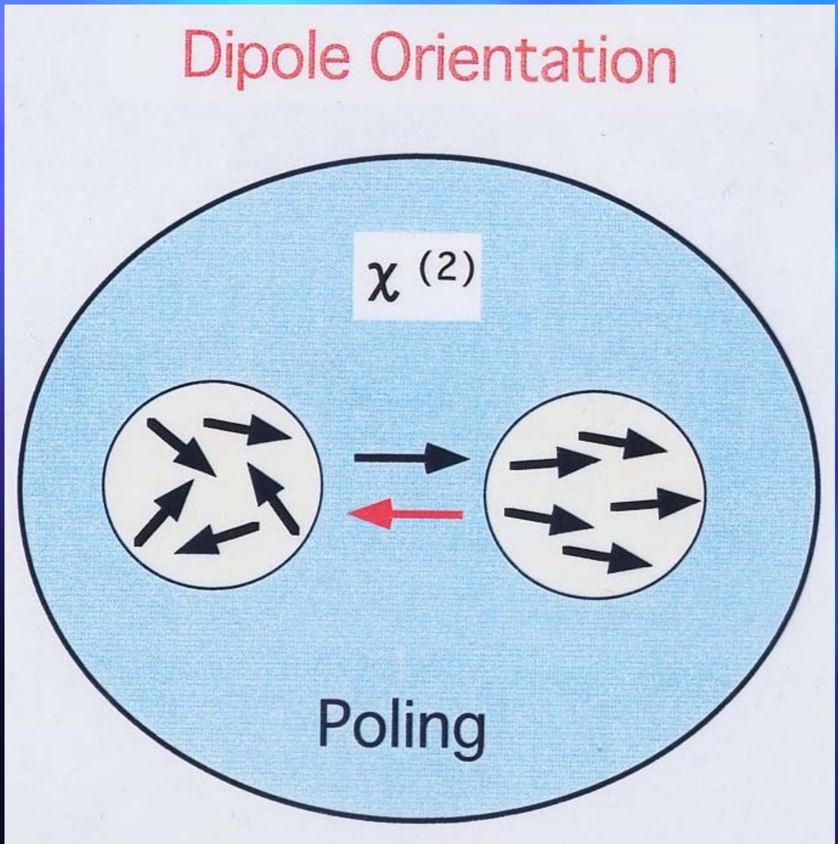


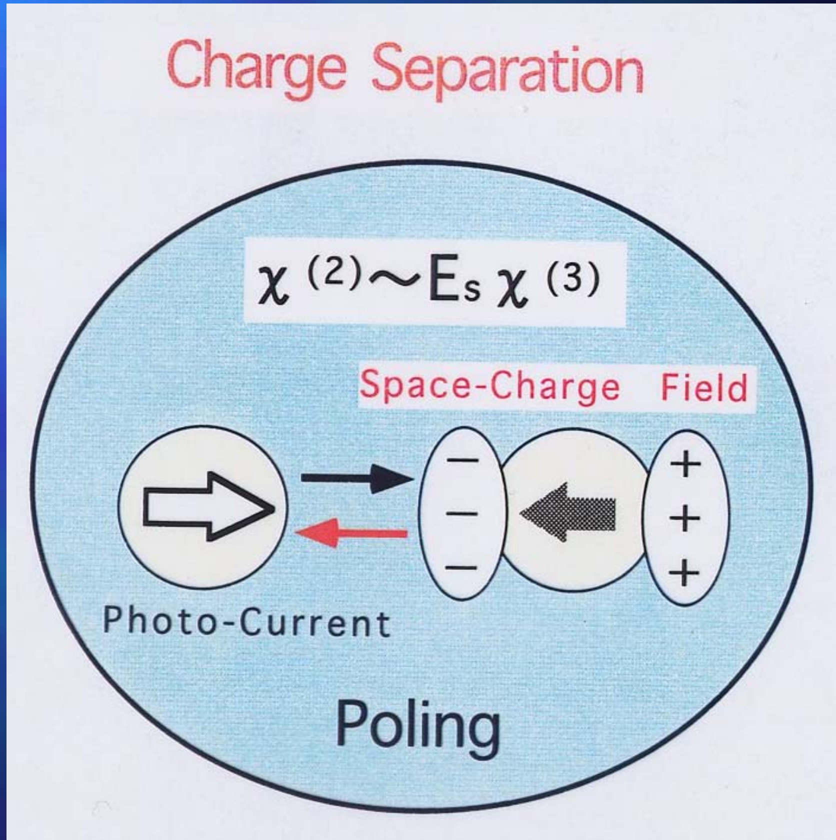
- -Larger $\chi^{(2)}$: **~10pm / V**
- -Periodic structure: $\chi^{(2)}$ **gratings**
- **-Degradation** → ***mechanism?***

Possible Origin of Induced $\chi^{(2)}$

Orientation of $\chi^{(2)}$ agents

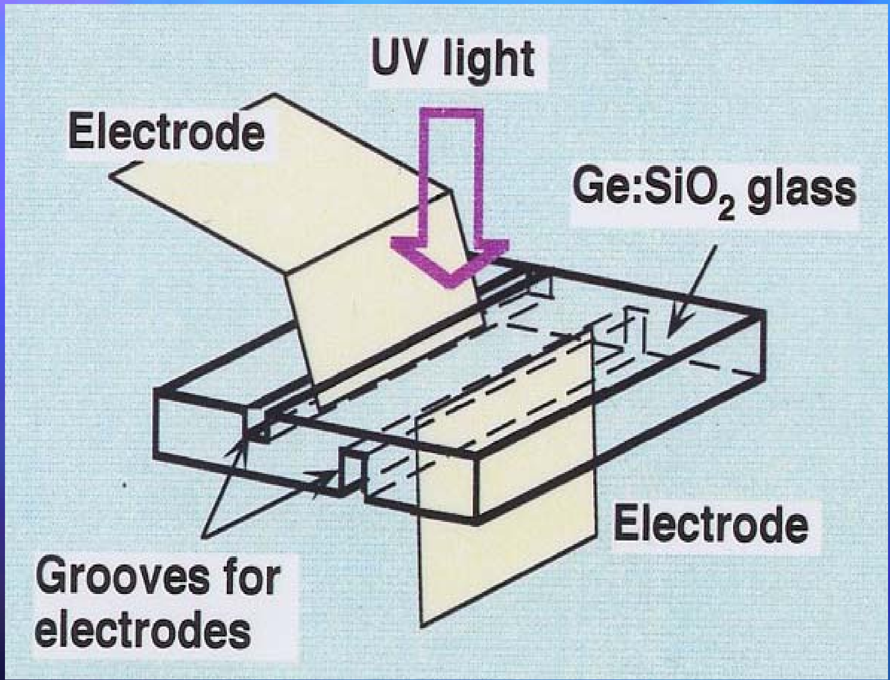
$\chi^{(2)}$ EE ~ $\chi^{(3)}$ EEE



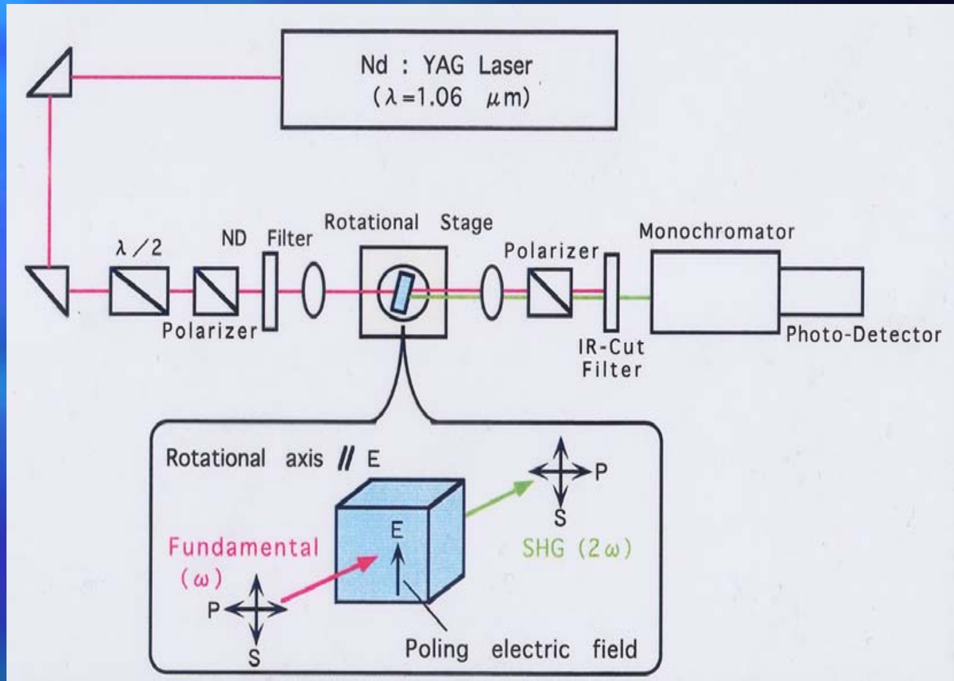


UV-Poling in Ge:SiO_2 Glass

UV-poling in bulk glass



Maker-fringe SHG measurement

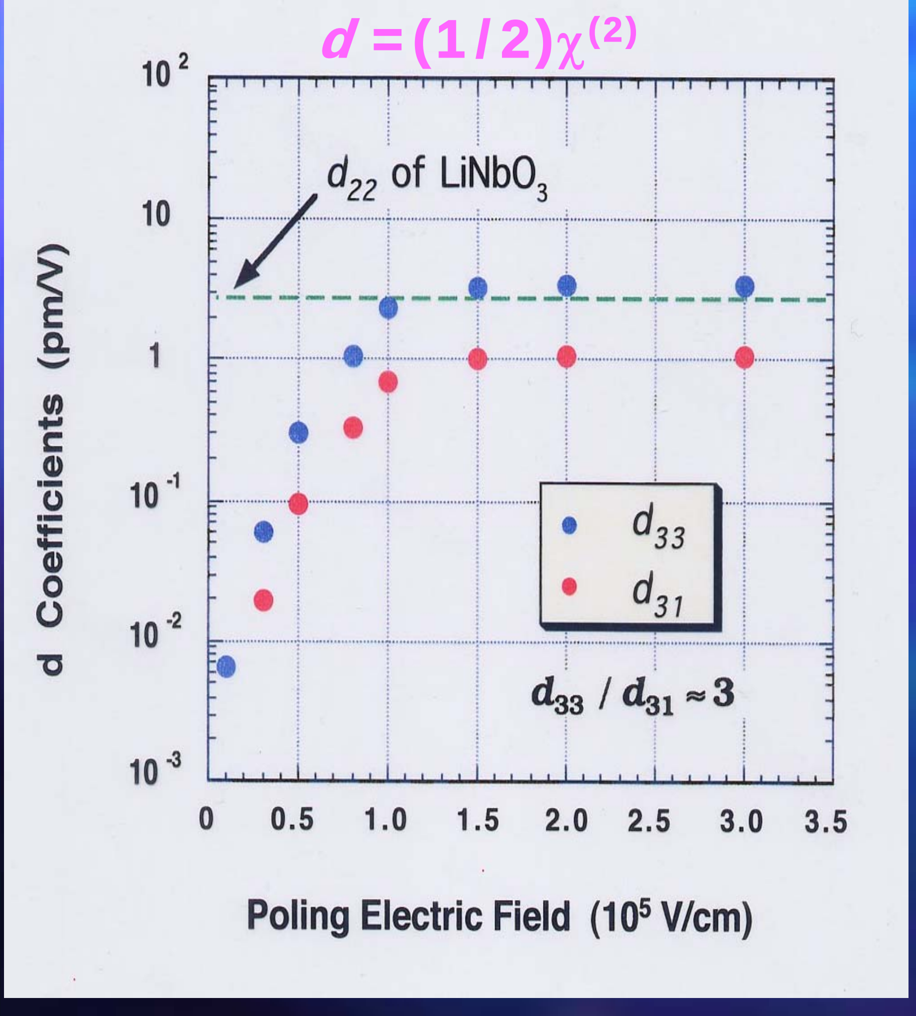


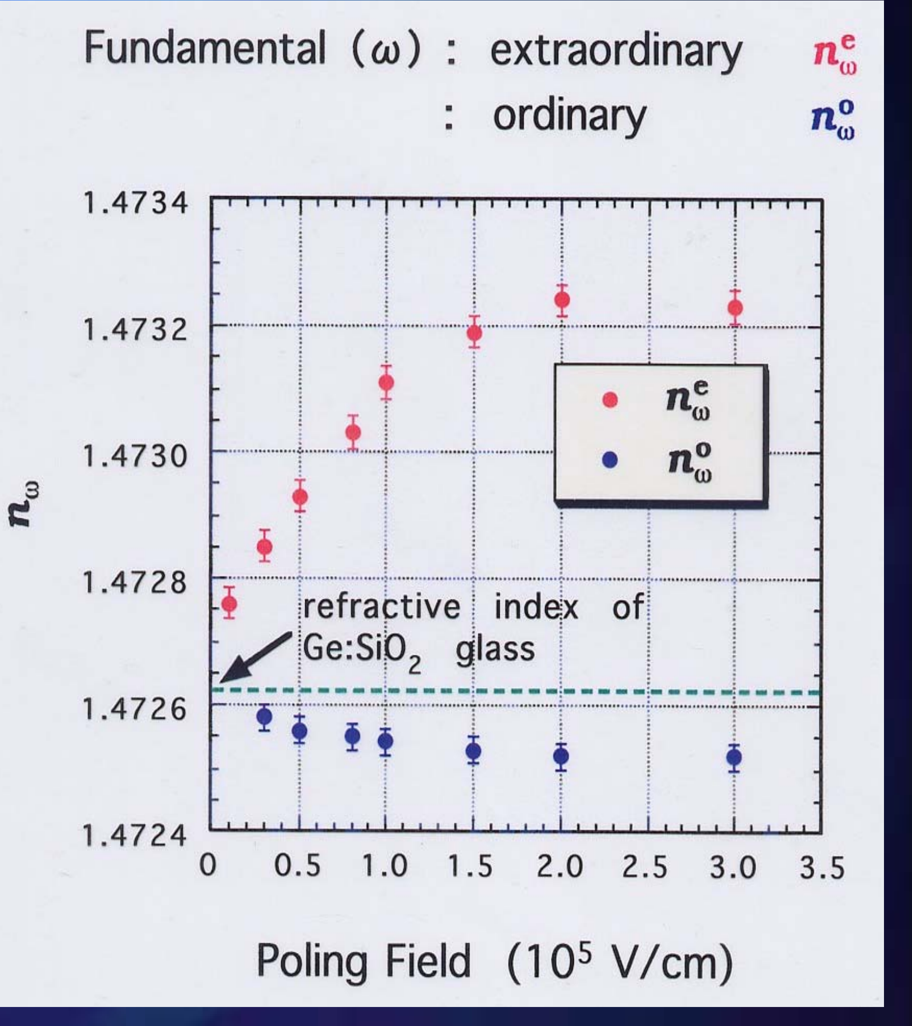
- VAD preforms: 15GeO_2-85SiO_2
- *E*-field: 0~3x10^5 V/cm
- UV-laser: 193 nm

- Quantitative evaluation of SHG *d* (χ) coefficients
- Values of d_{33}, d_{31}
- Refractive index: n_e, n_o

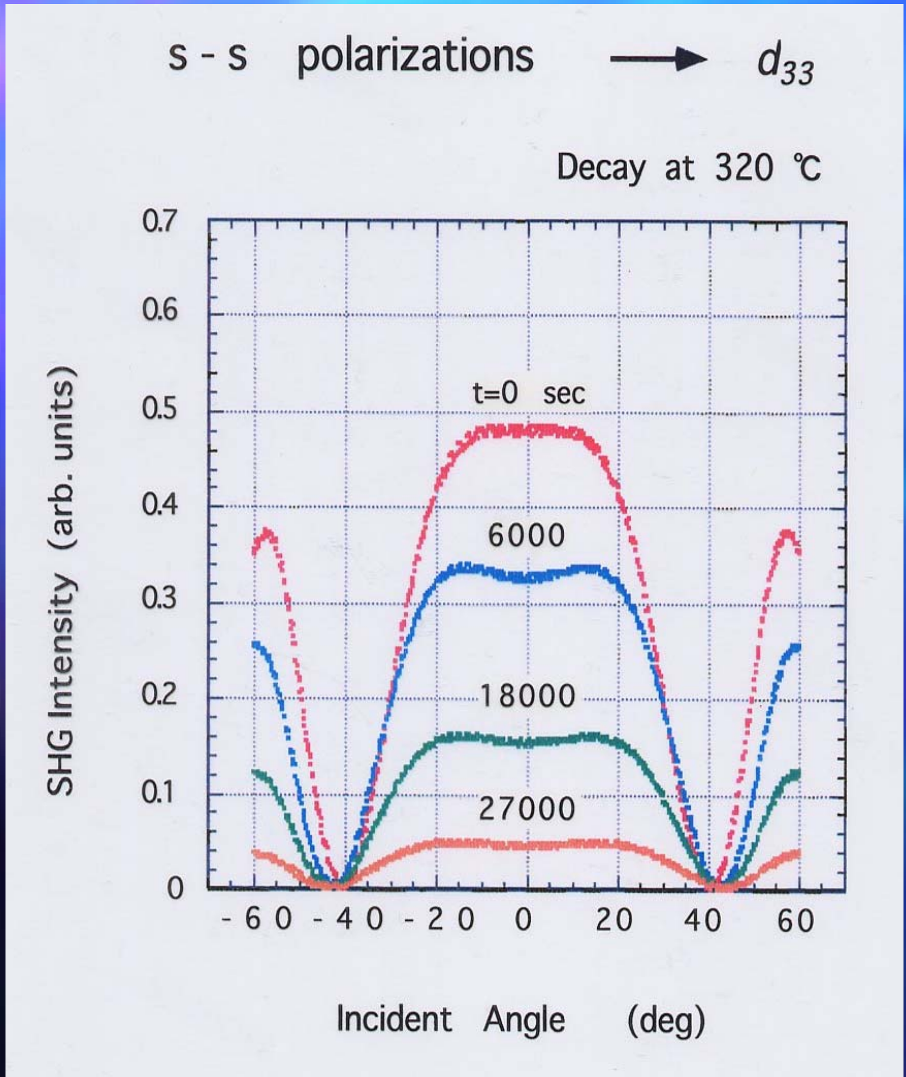
Creation of $\chi^{(2)}$ in UV-Poled Glass

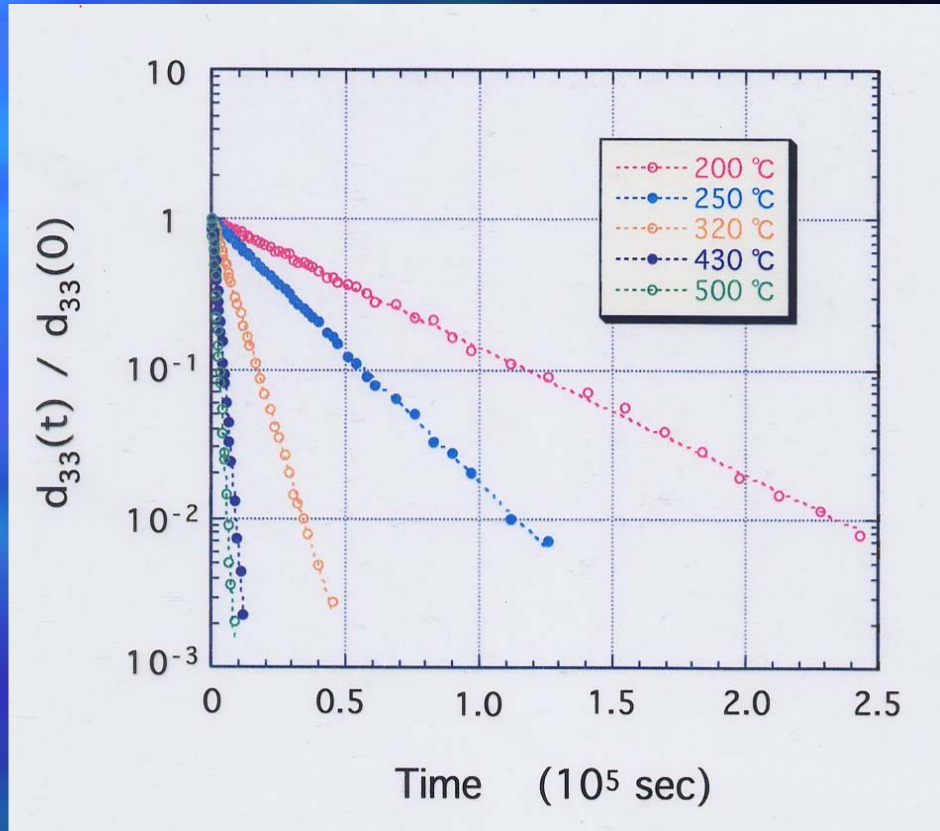
UV-poling electric field dependences in Ge-doped SiO_2





Decay Behaviors of Induced $\chi^{(2)}$



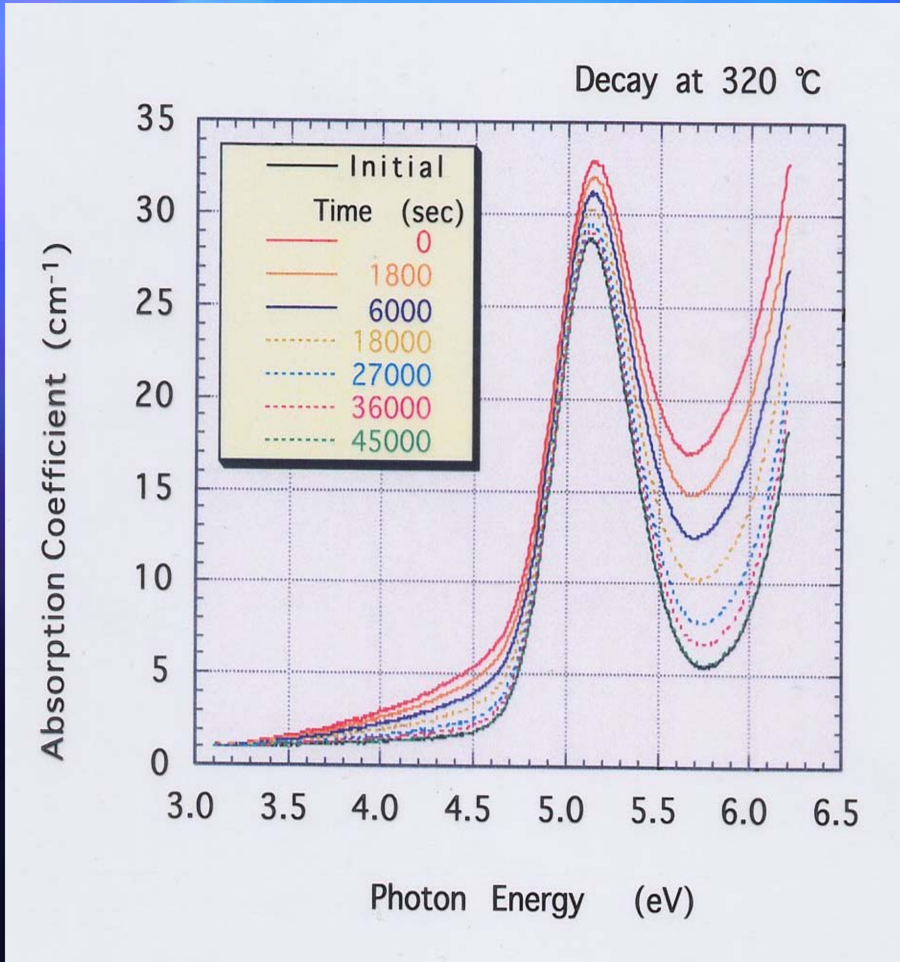


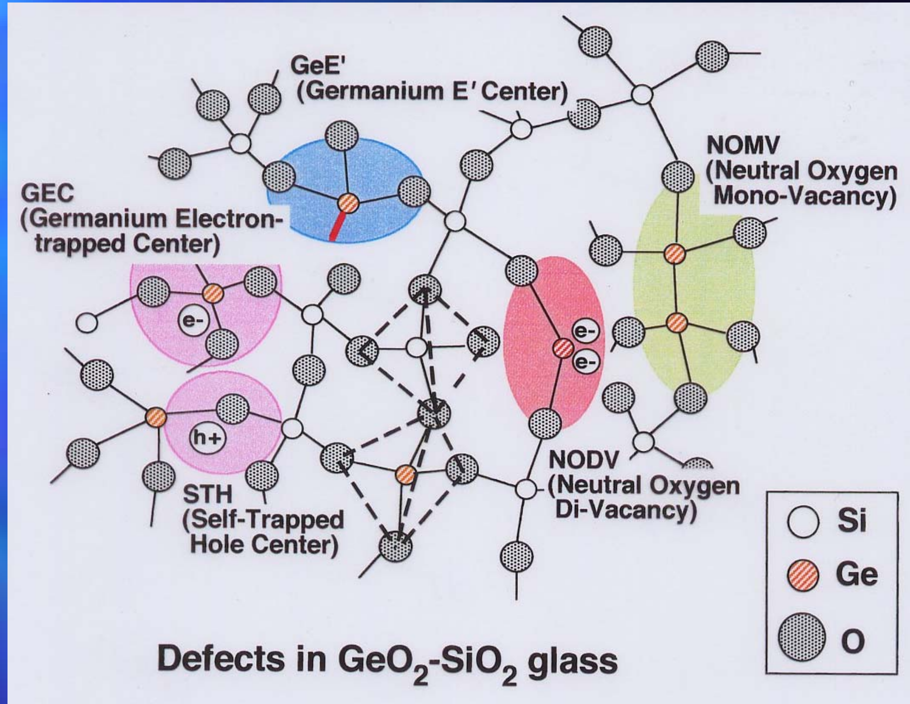
-$\chi^{(2)}$ disappearance

-single-expo. decay?

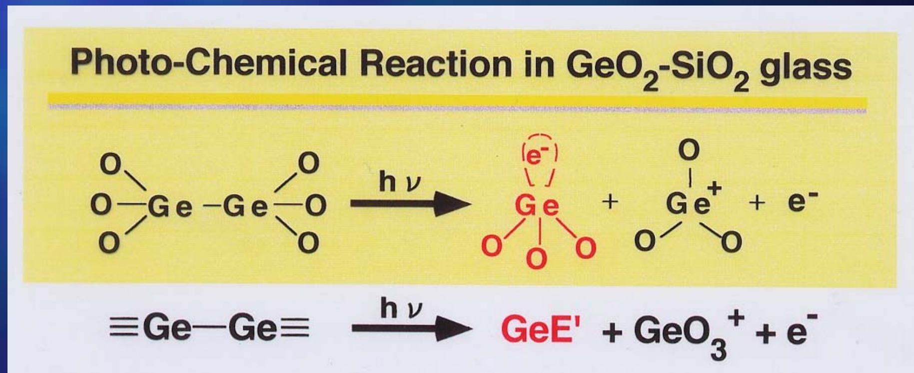
Quantitative Analysis of Decay (1)

Absorption Spectra and defects in Ge-doped SiO_2 Glass



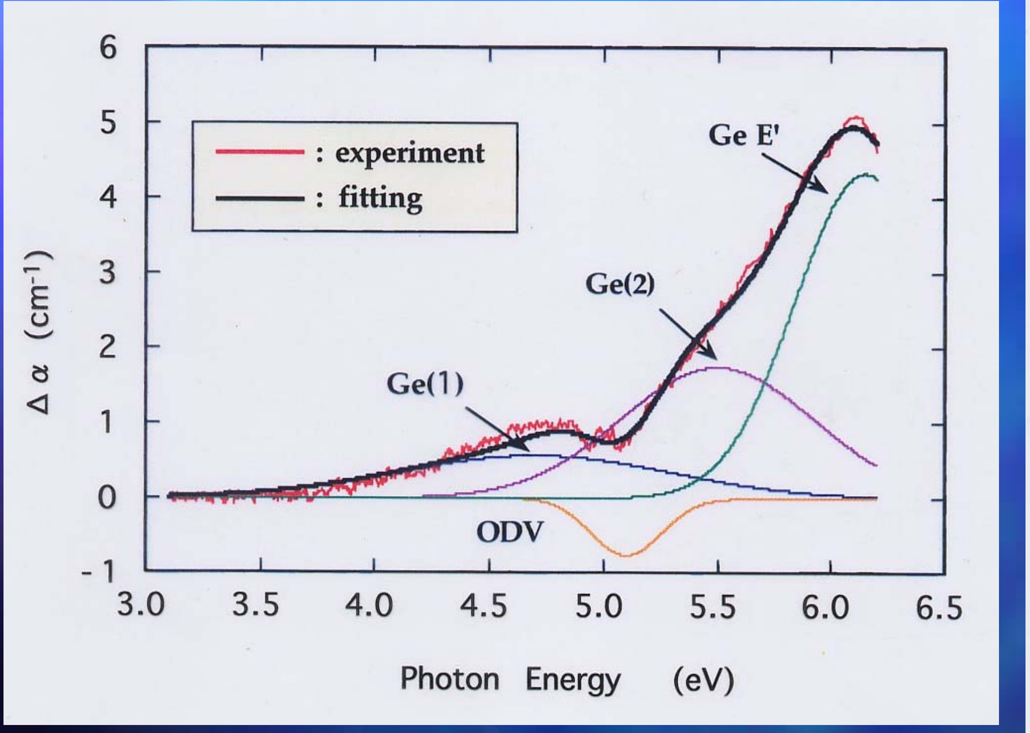


Defects in GeO_2-SiO_2 glass

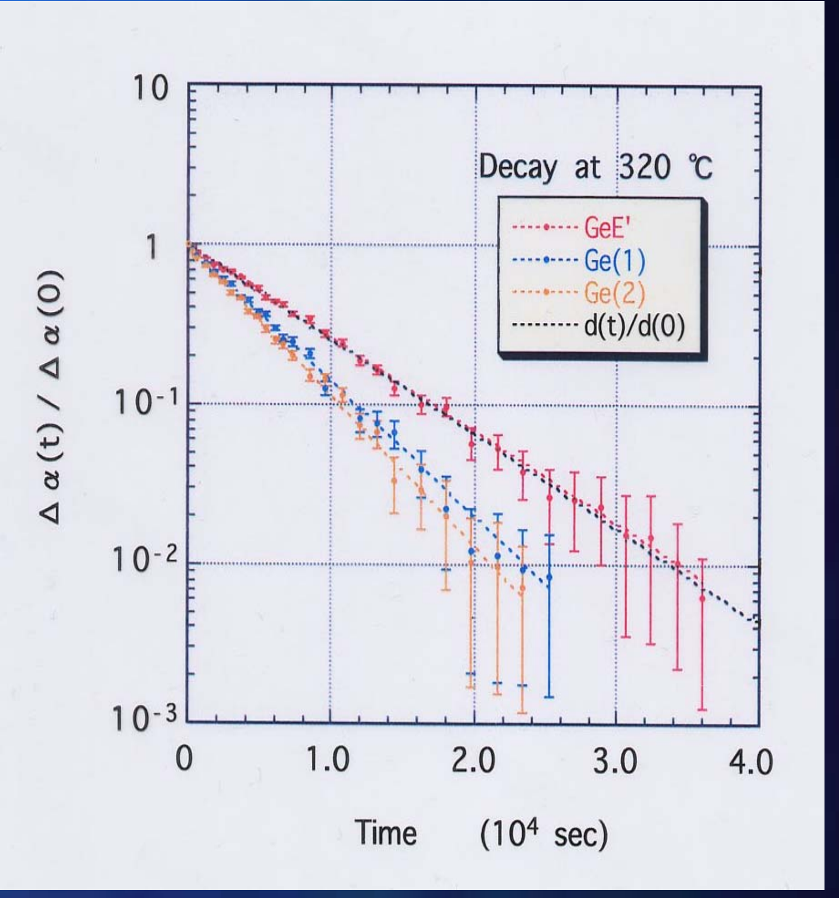


Quantitative Analysis of Decay (2)

Deconvolution of Δα

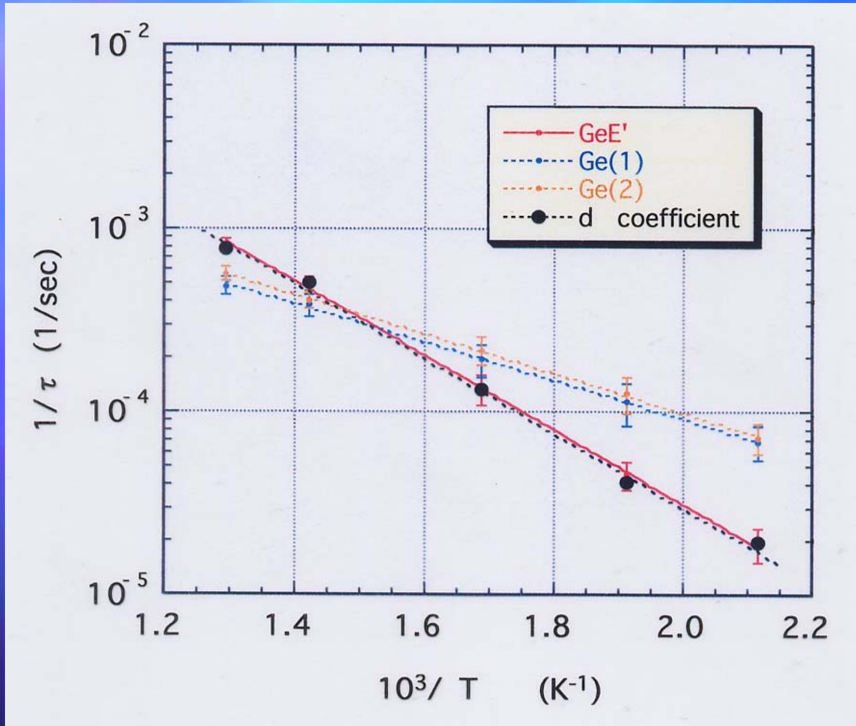


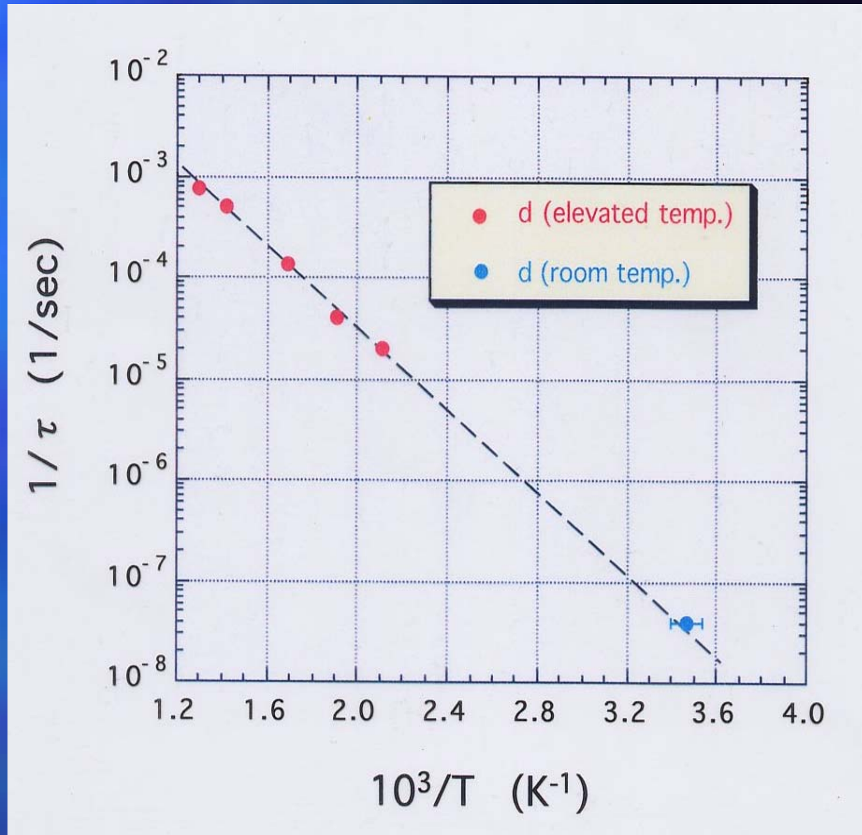
Decay of Δα



$\chi^{(2)}$ decay is similar to GeE' !

Decay Time Constant of Induced $\chi^{(2)}$





	activation energy (eV)
d coeff.	0.41±0.05
GeE'	0.40±0.10
Ge(1)	0.21±0.09
Ge(2)	0.22±0.09

Decay time constant of $\chi^{(2)}$ induced in UV-poled glass

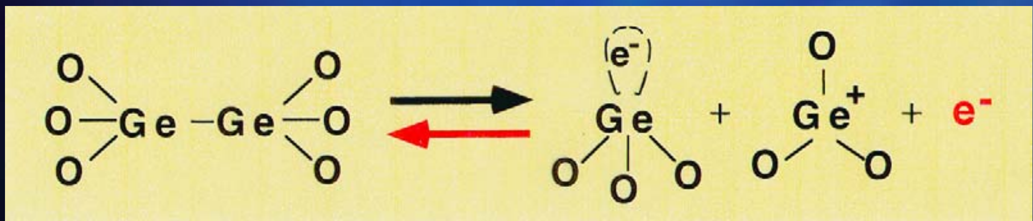
~280 days at RT

Mechanism of $\chi^{(2)}$ Decay

Comparison of activation energies

	Activation Energy (eV)
Decay of d Coefficient	
bulk (untreated)	• 0.41±0.05
bulk (heat treated)	0.38±0.05
Dark Conductivity	
bulk (untreated)	• 0.44±0.05
bulk (heat treated)	0.37±0.05
Defects	
Ge-E'	• 0.40±0.10
GEC*	0.21±0.09

*Ge-related electron trapped centers



Values of E_a

$\chi^{(2)}$ decay and GeE'

~0.4 eV

Dark conductivity

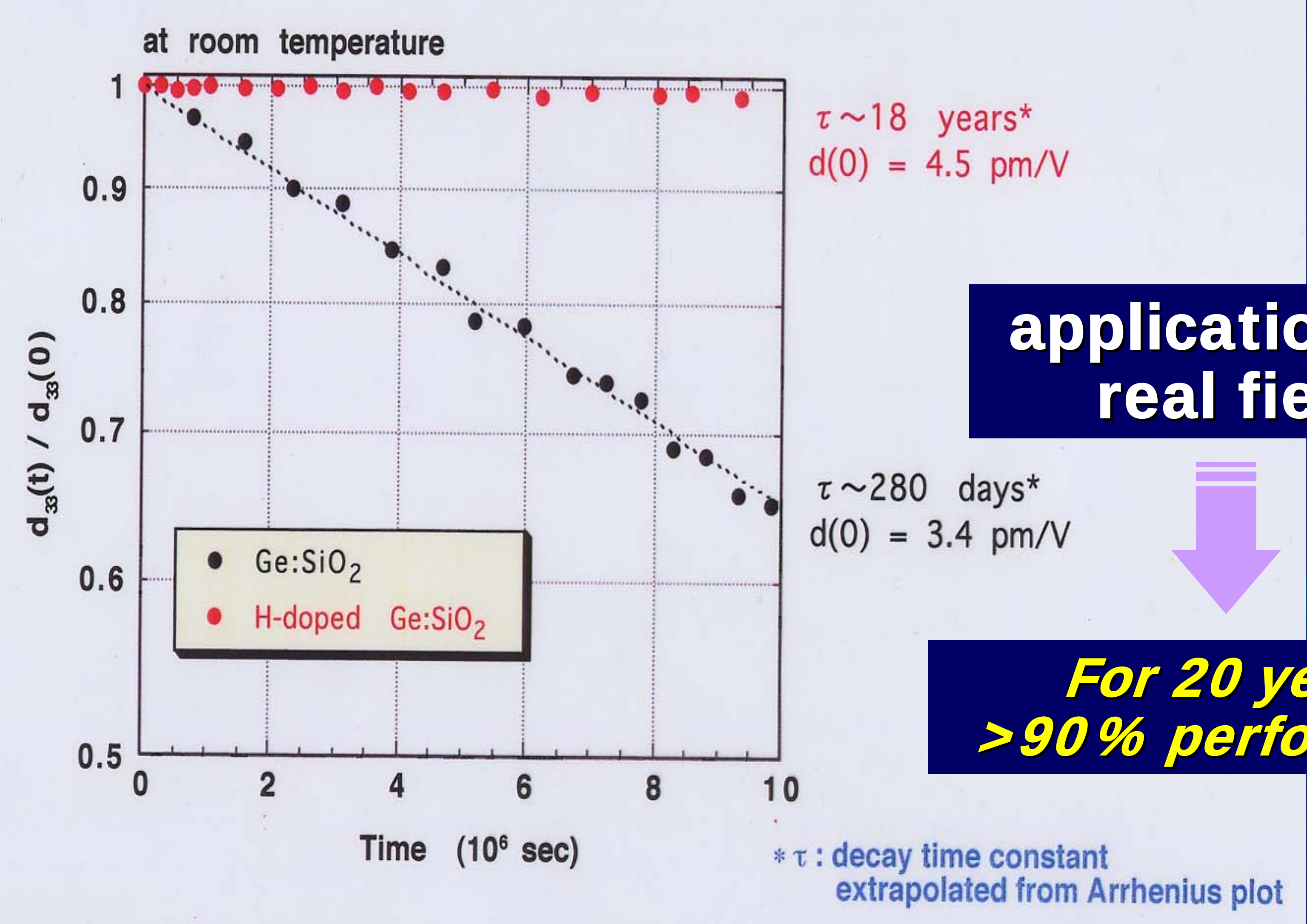
~0.4 eV

Introduction of electron scavengers?

For long-term stability

Hydrogen doping

Achievement of Stable $\chi^{(2)}$



application to real field

For 20 years, >90% performance

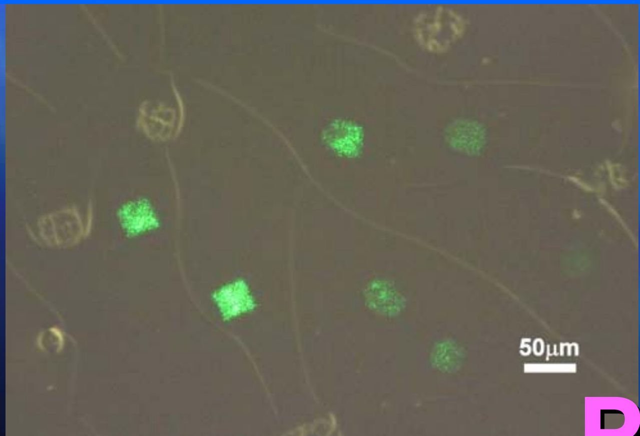
Origin and Decay of $\chi^{(2)}$ in UV-Poled Glass

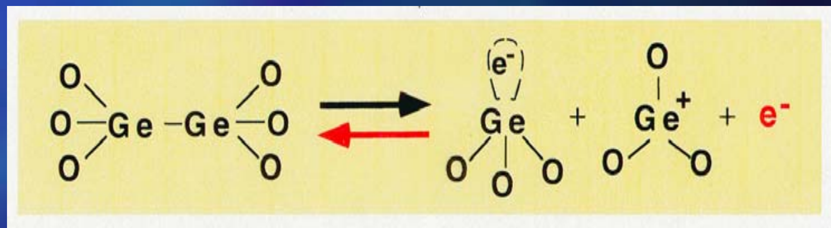
Effective $\chi^{(2)}$ through third-order nonlinearity

$$\chi^{(2)} \sim \chi^{(3)}\, E_{sc}$$

$\chi^{(3)}$ susceptibility: increased by crystallization

Esc : space-charge field caused by defect formation



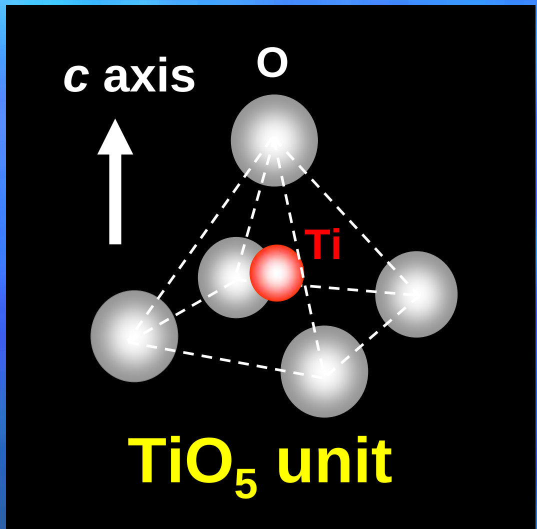


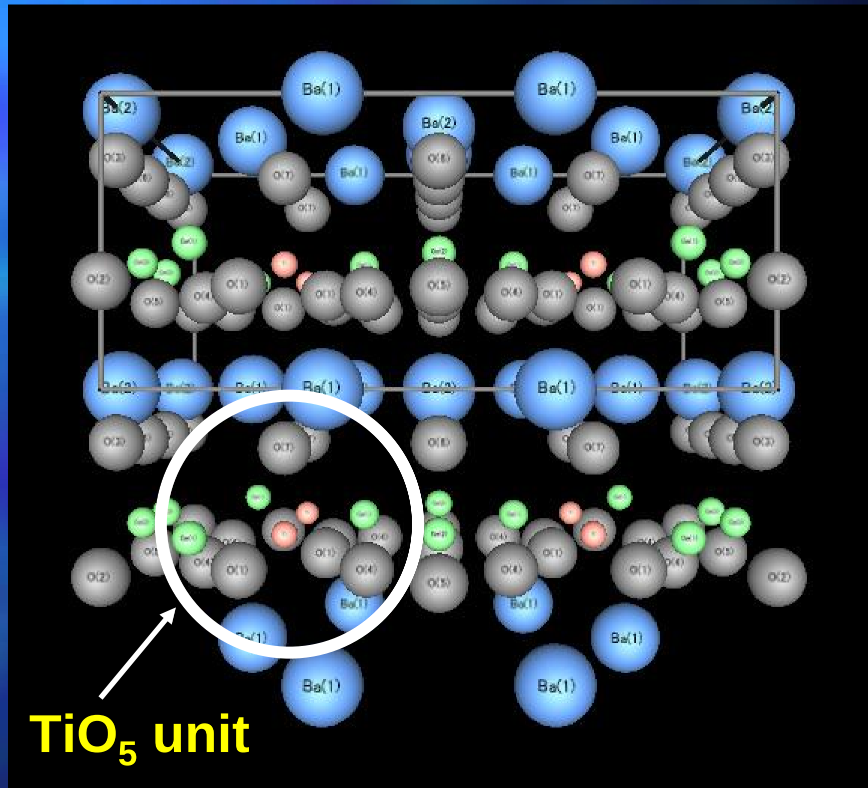
Permanent $\chi^{(2)}$?

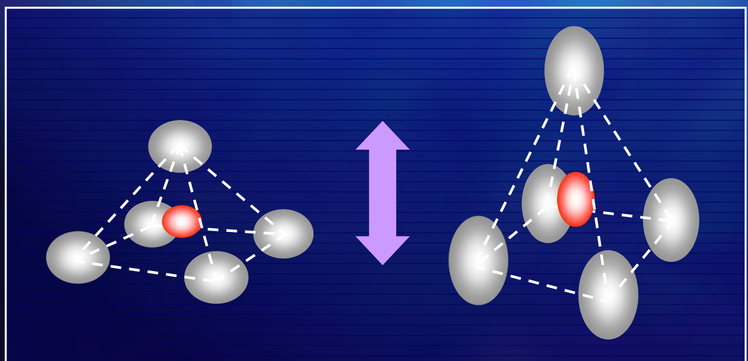
$Ba_2TiGe_2O_8$ (BTG)

Fresnoite Crystalline Structure

Origin of P_s (spontaneous polarization)

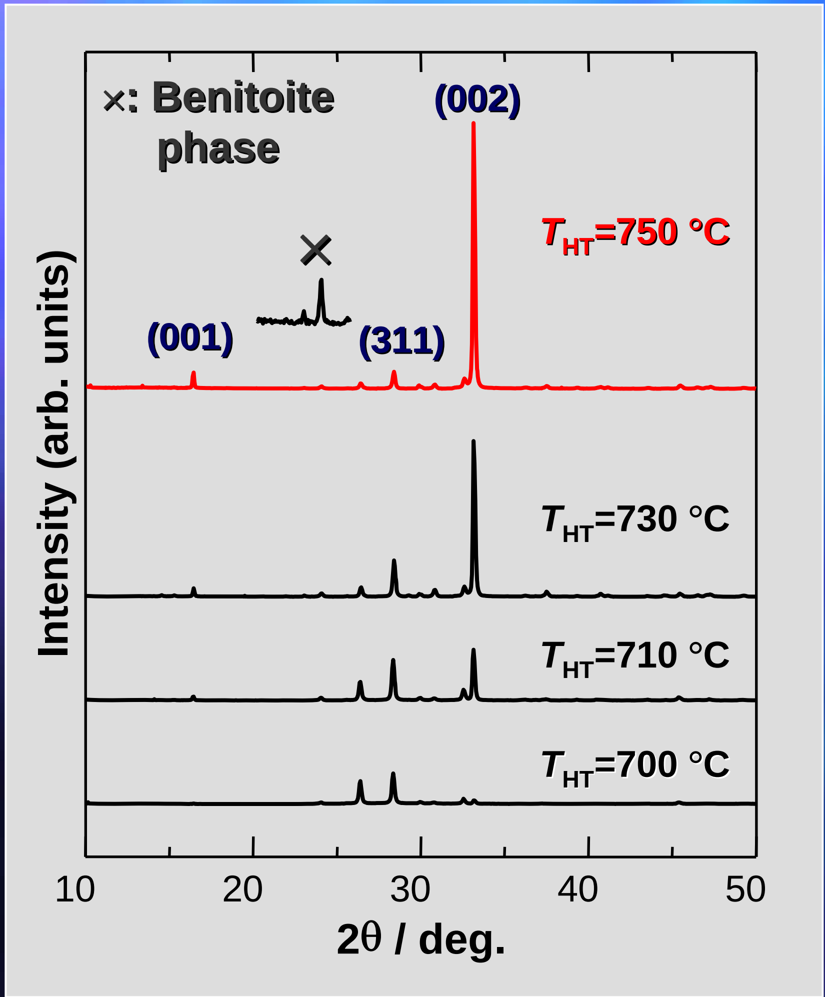


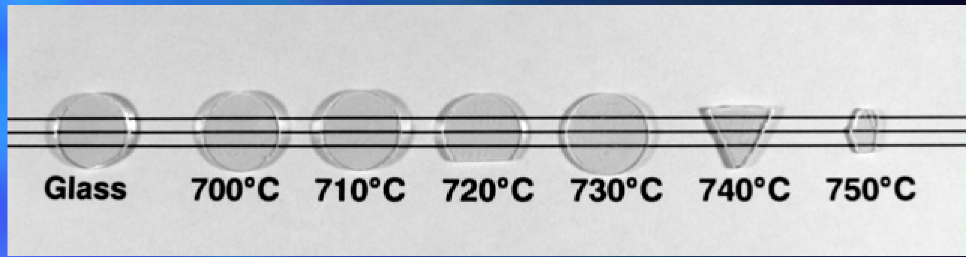


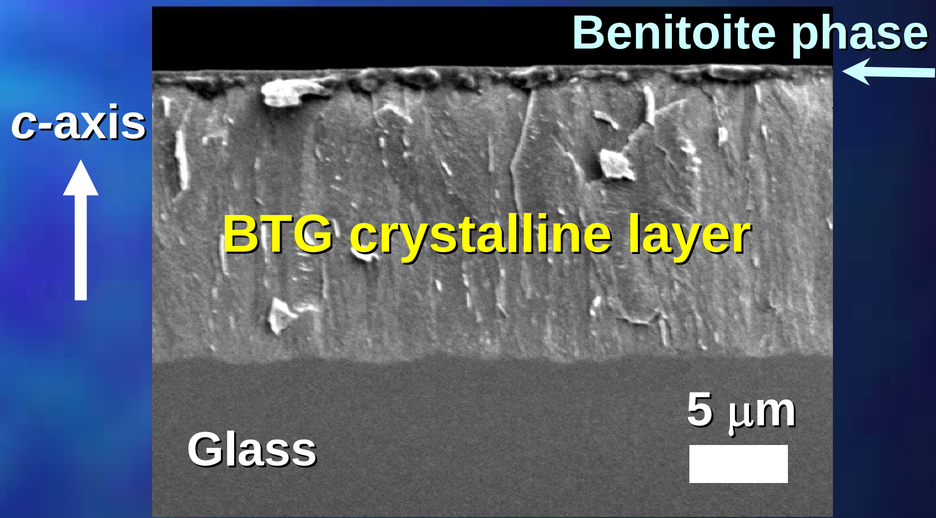


Novel Crystallized Glass－BTG

Surface Crystallization and Orientation



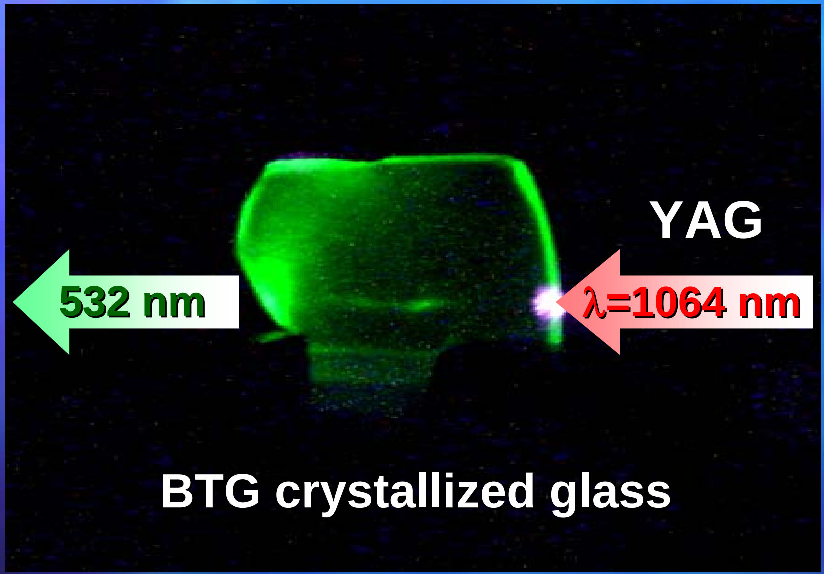




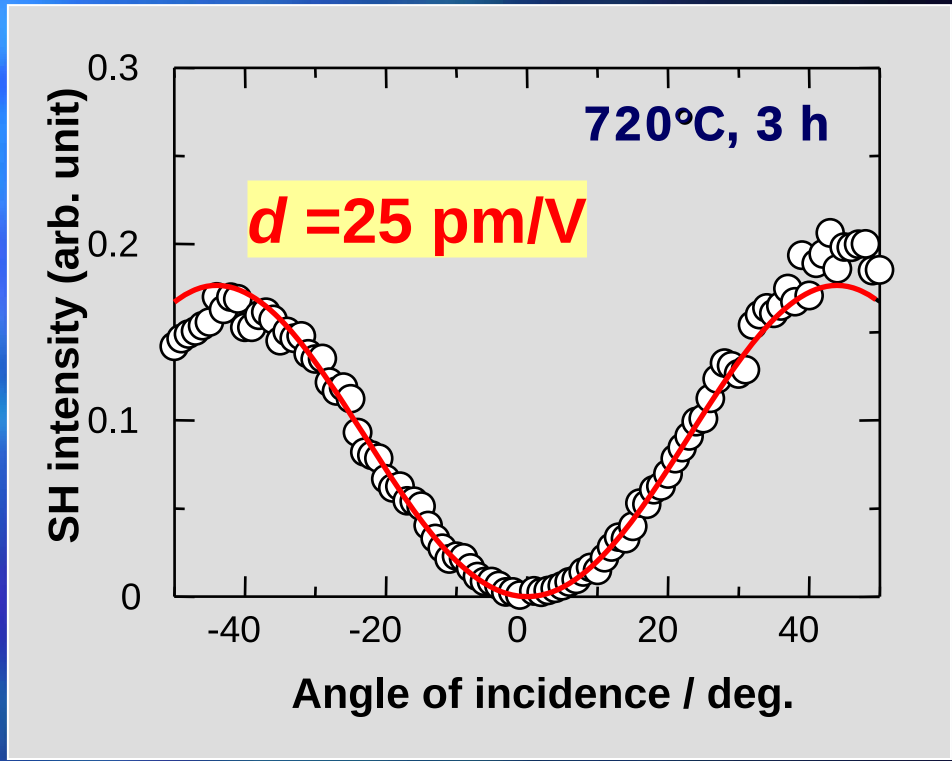
Stoichiometric composition

2nd-Order Nonlinearity in BTG

BTG55: $30BaO_2$-$15TiO_2$-$55GeO_2$



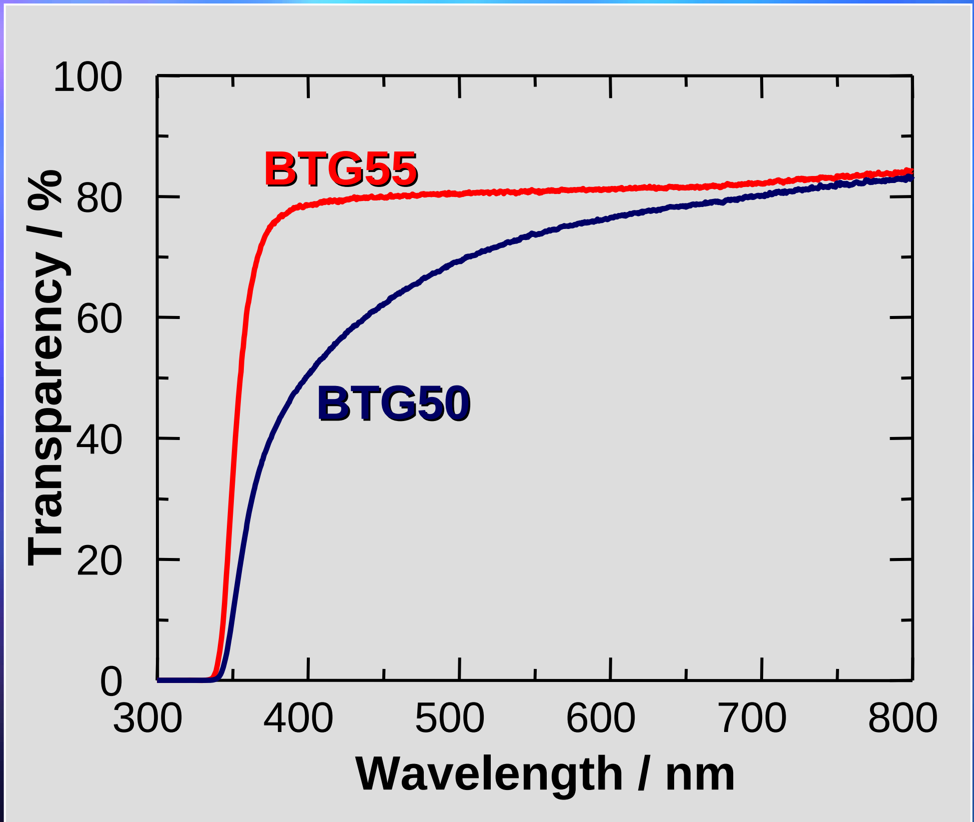
Appl. Phys. Lett., 81, 223(2002).

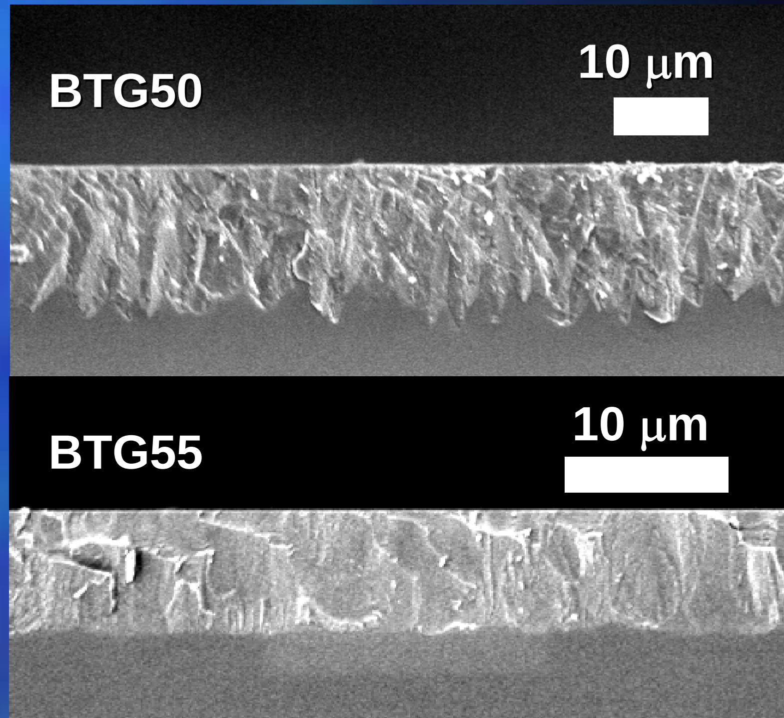


Maker fringe measurement:

The largest *d*-value in glass ever reported

Optical Absorption and Microstructure of BTG55 and BTG50





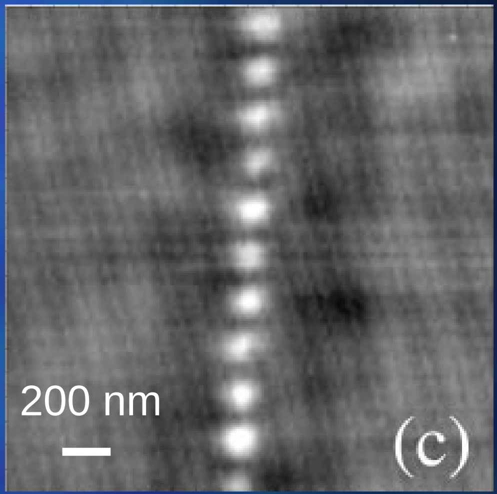
Crystalline layer of BTG55 is more dense and homogeneous than those of BTG50.

Plasmonics

Surface plasmon locallized in metal nano-particles

J. R. Krenn (2001)

-electron beam lithography (EBL)
-ITO doped glass substrates with electric conductivity for EBL
-gold nano-particles with 100 nm diameter and 40 nm height for a plasmon resonance wavelength of about 630 nm
-plasmon coupling observed by photon scanning tunnelling microscope (PSTM)

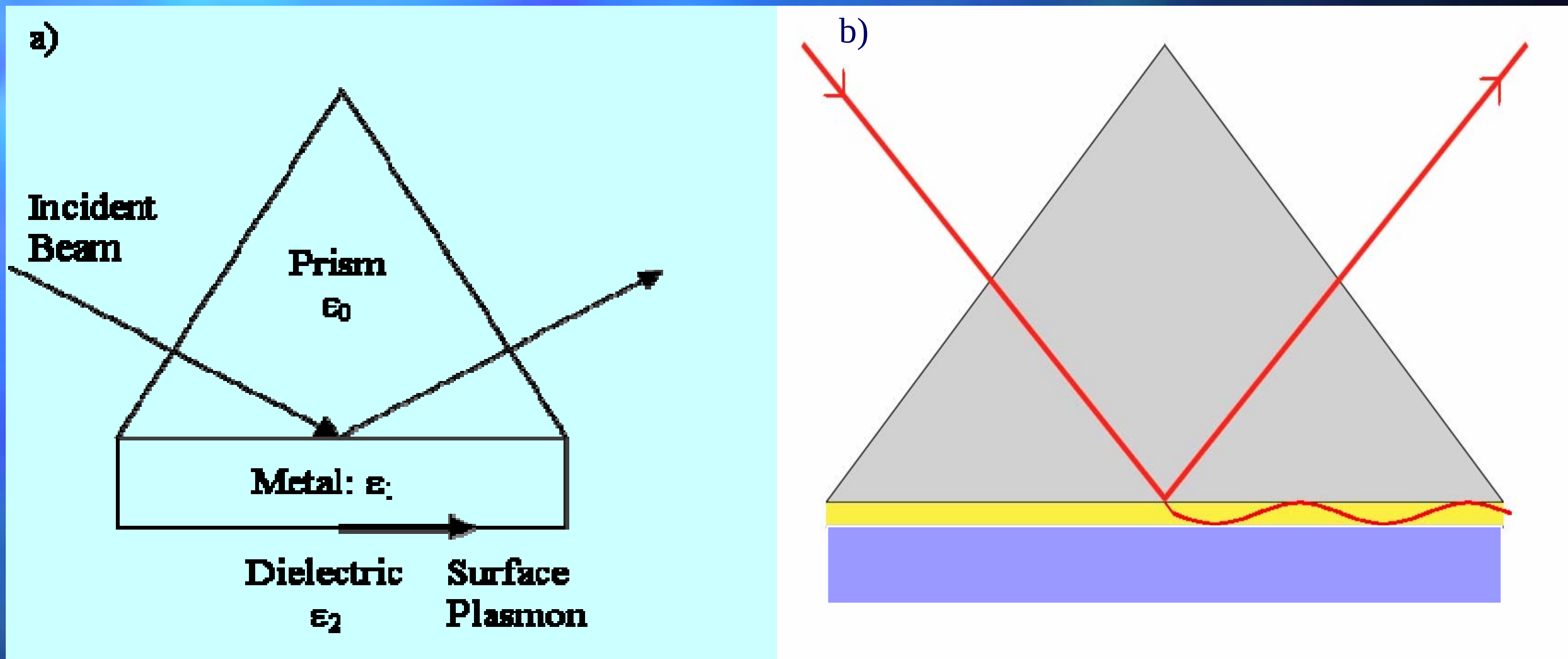


Optical intensity image of Au nano-particles ordering in glass substrate

J. of Microscopy, 202, (2001) 122

Suraface Plasmon (SP)

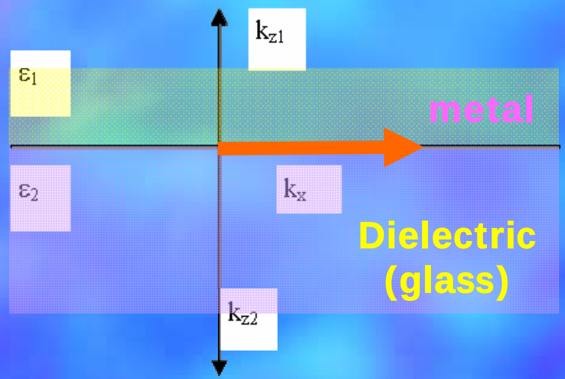
1. Excitation of SP by photon coupling



a) Kretschmann configuration and b) ray tracing of an Attenuated Total Reflection (ATR) setup for coupling surface plasmons.
In the case, the surface plasmon propagates along the metal/dielectric interface.

Suraface Plasmon (SP)

2. Dispersion relationship for SP



Wave number of SP: k_x

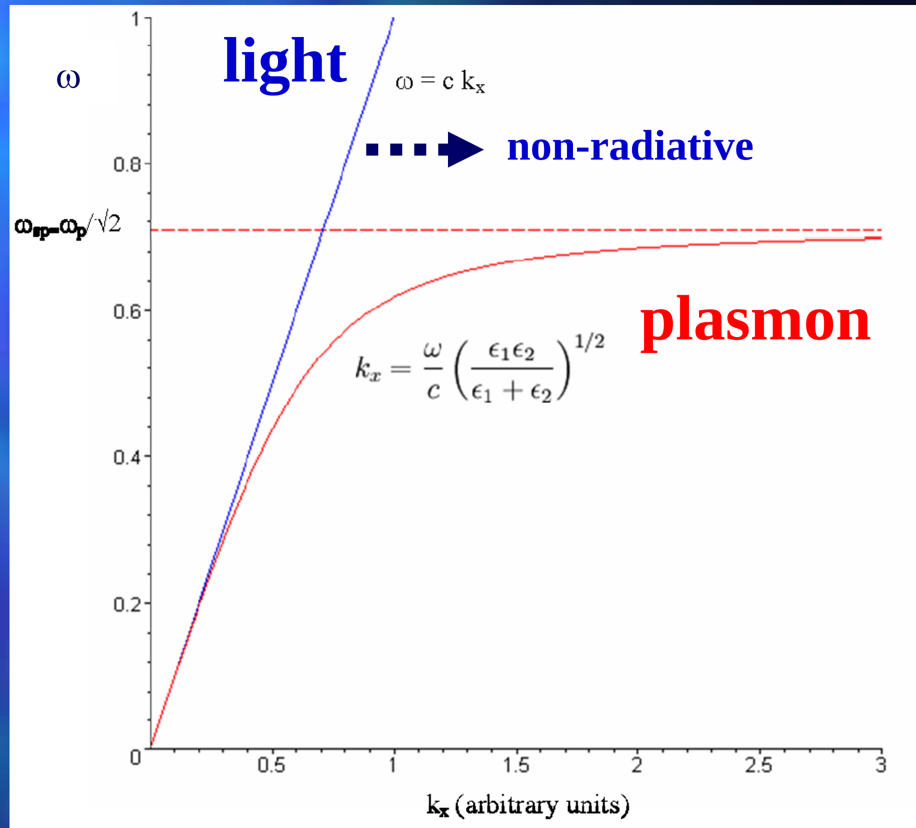
Dielectric constants (relative): ε_1 and ε_2 for metal and dielectric, respectively.

$$k_x = \frac{\omega}{c}\left(\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}\right)^{1/2}$$

c : speed of light, ω : frequency of the wave

Since $\varepsilon_1 < 0$ in metal, for the solution of k_x (plasmon),

$\varepsilon_1(\omega) < -\varepsilon_2$, below ω_{sp}



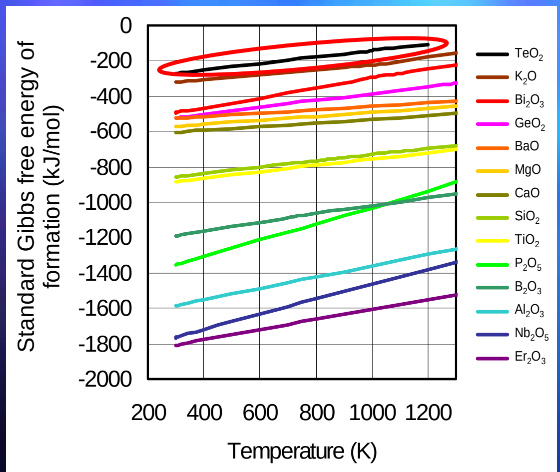
Dispersion curve for surface plasmons. At low *k*, the surface plasmon curve (red) approaches the photon curve (blue).

Laser-Induced Structure Ordering

Tellurite-based glasses

- Nano-crystallization by laser heating
- Selective crystallization of metal Te?
- Large nonlinearity: $d \sim 30d$ ($LiNbO_3$)

  $\chi^{(3)} \sim 10\chi^{(3)}$ (Au)



$KNbO_3$-TeO_2 glass

Periodic Structure with PM

XeCl excimer laser (λ=308nm)

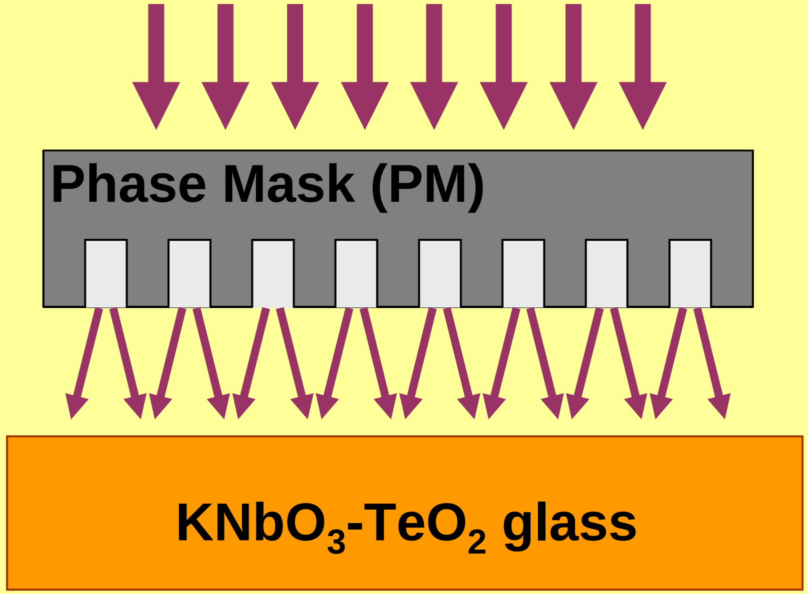
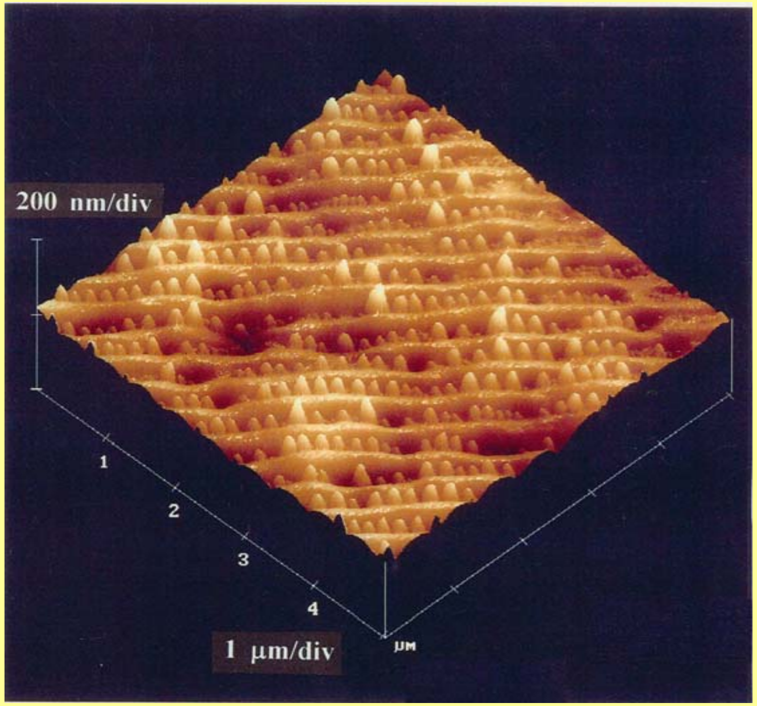


Photo-Induced Nano-Crystallization by UV-Laser Irradiations

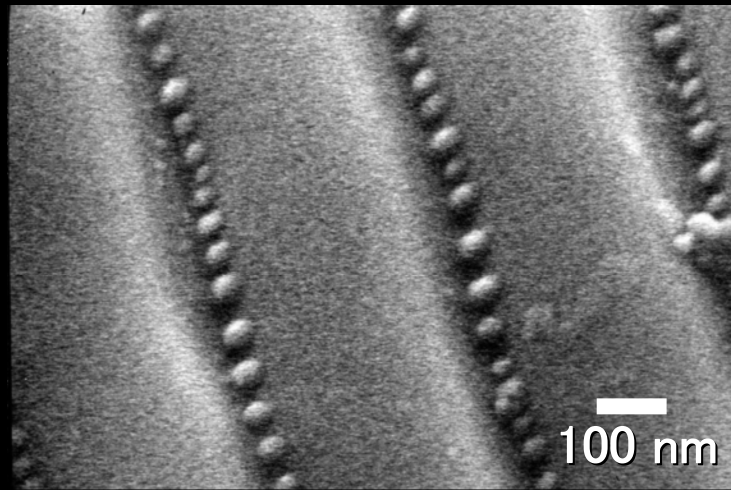
Periodic Structures of Nano-Particles 2

Structure Ordering in Glass

AFM image (enlarged)



SEM image



ordered structure of nano-particles

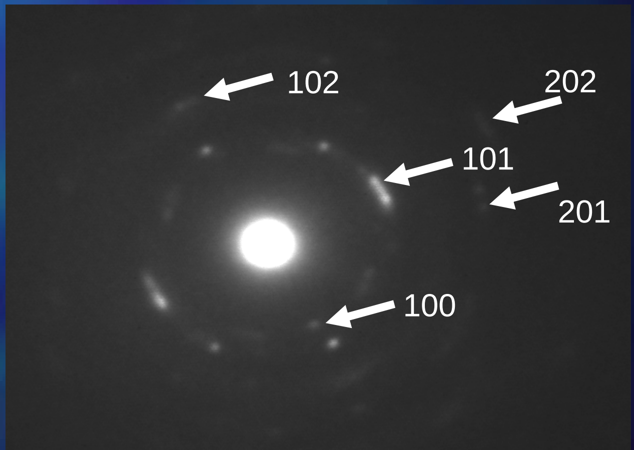
TEM Images of Surface Cross-Section

UV-Irradiation

-Creation of nano-particles with ~100 nm diameter
-Laser intensity dependence of nano-particles density
-Te metal confirmed by electron diffraction pattern

Metallic Nano-Structures on Glass Surface

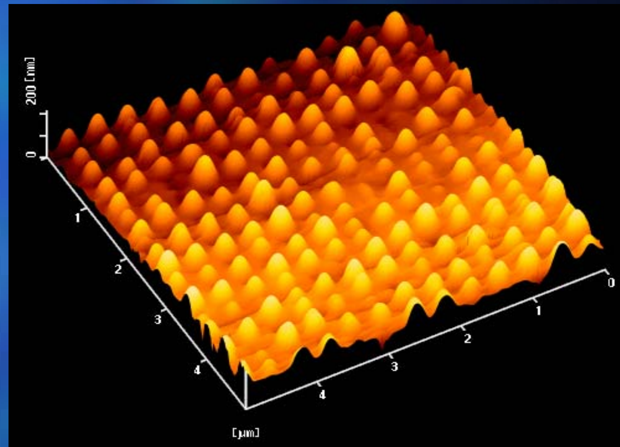
Plasmonic Glass

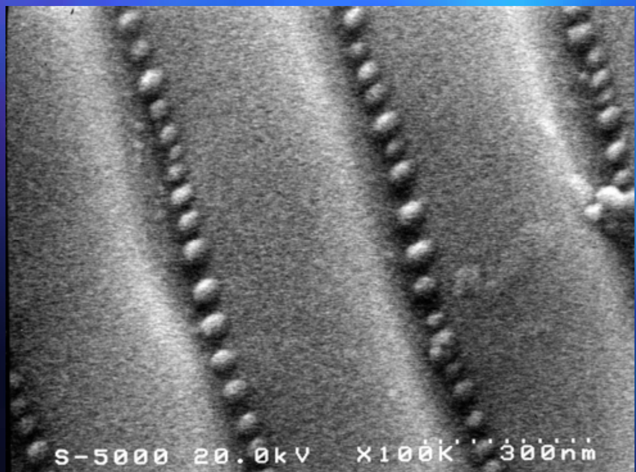


Plasmonic Glass for Nano-Circuit

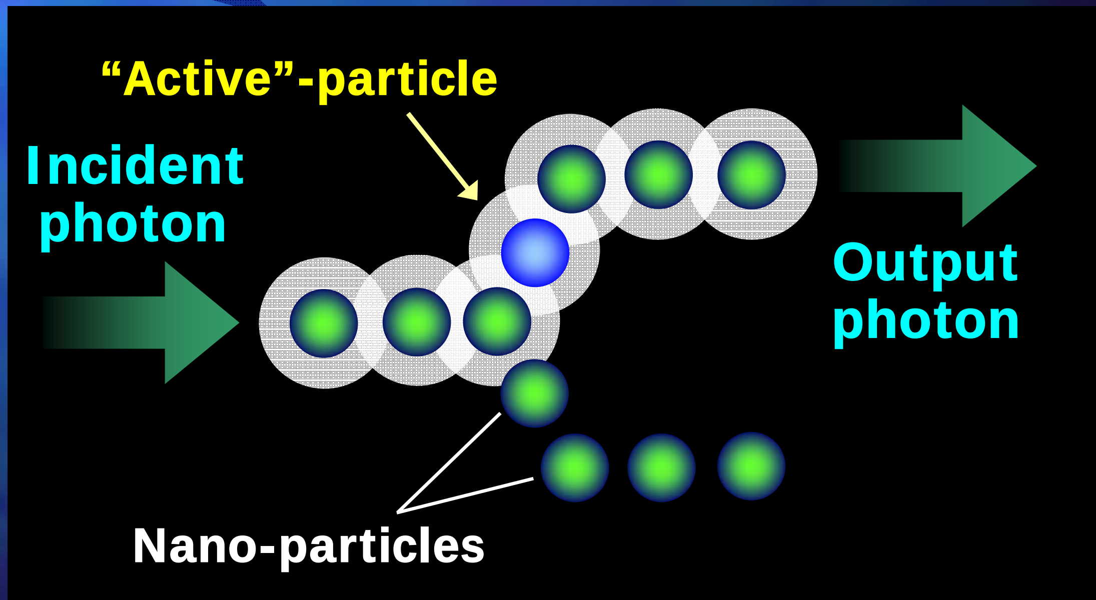
Photo-Induced Nano-Particles Structure

- Metal nano-particles on glass
- Physics for formation
- Design and control of particles
- Nano-photonic circuits

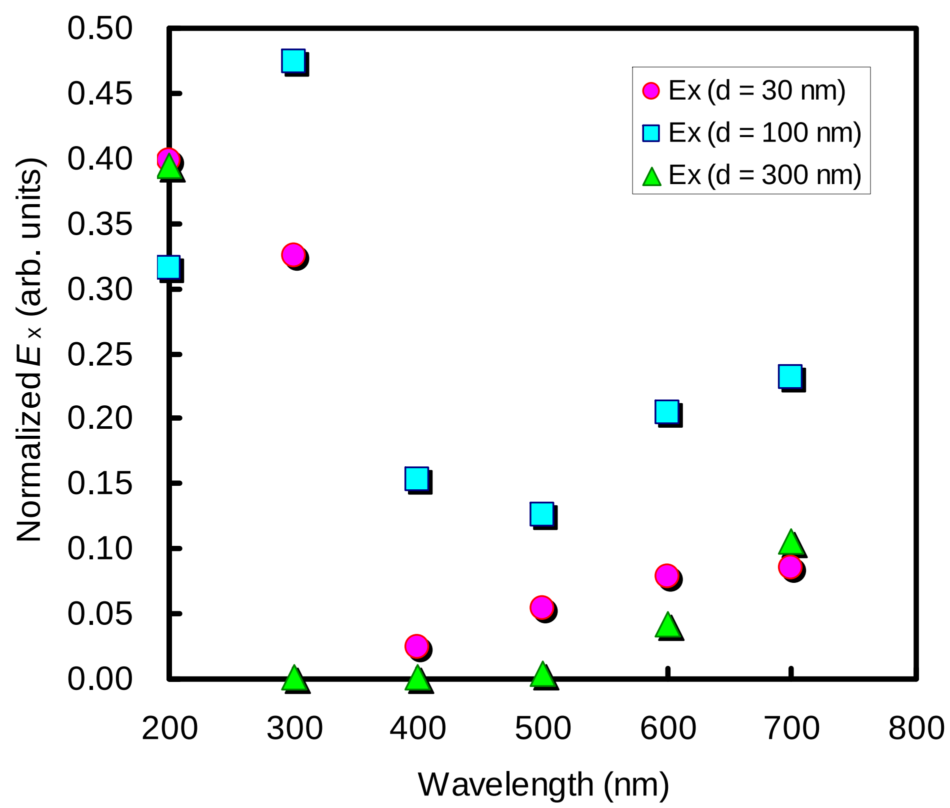


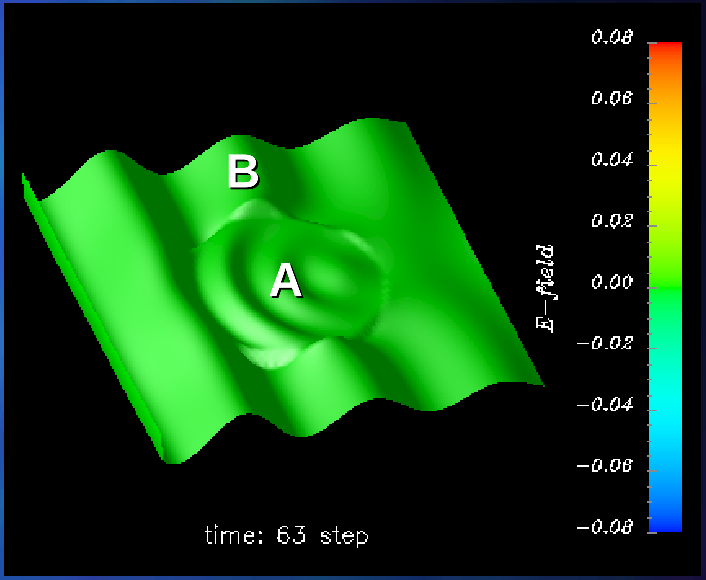


Ordered Nano-Particle Structure



Change of E-field Intensity （FDTD）





$$\text{Normalized Ex} = \frac{\text{E-field: A}}{\text{E-field: B}}$$

Low Degradation of E-field in 100 nm

SUMMARY

Controlling Light with Nonlinear Optical Glasses and Plasmonic Glasses

- **Developments of new nonlinear optical glasses for EO photonic devices**

  Fiber-Type Devices

  for Signal Processing in Optical Communication

- **Formation of UV-laser induced metallic nano-particle structures on glass surface**

  Plasmonic Glass

  for Propagation / Localization of Light

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