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Lecture 1, Part 2: Controlling light with nonlinear optical glasses and plasmonic glasses - Plasmonics

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*2008, Jan 7
All-Paid US-Japan Winter School
on
New Functionalities in Glass*



TOHOKU
UNIVERSITY

Photonic Glass

*Controlling Light with Nonlinear Optical
Glasses and Plasmonic Glasses*

Takumi FUJIWARA

*Tohoku University
Department of Applied Physics
Optical Materials and Sciences Lab.*

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

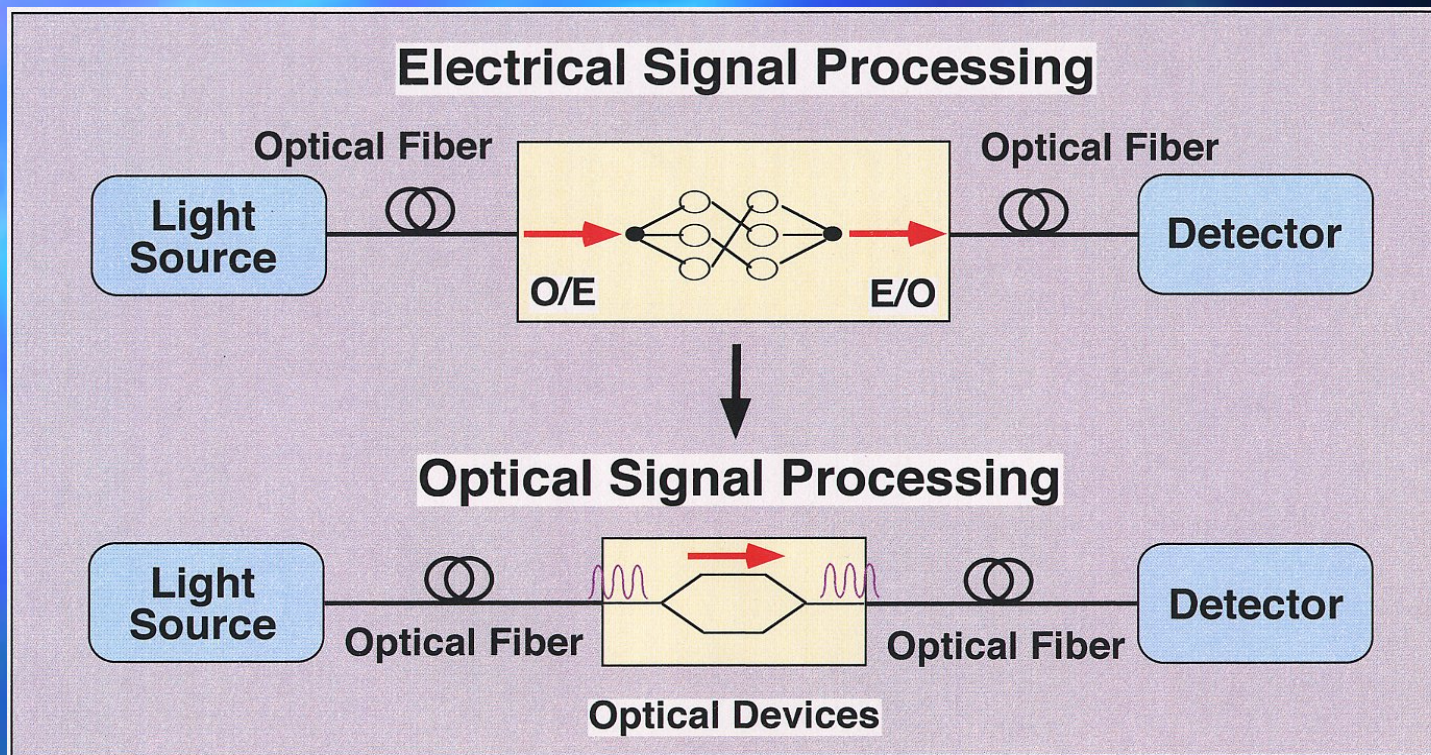
Novel nonlinear “glass materials” for photonic applications

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
driven by *Second-Order Optical Nonlinearity*

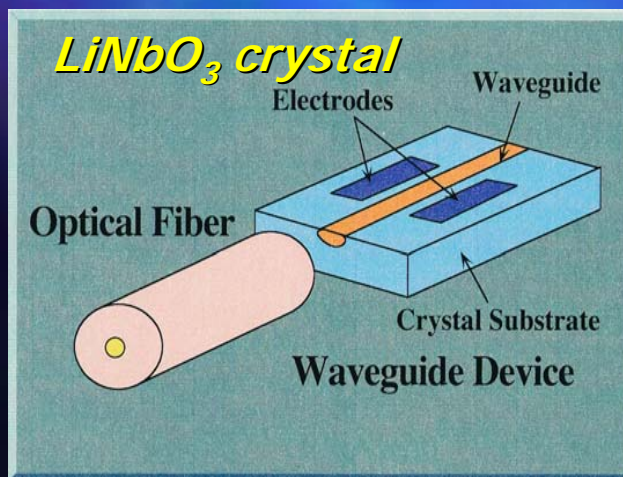
Second-Order Optical Nonlinearity in Glass

2nd-order optical nonlinearity

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

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

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



Permanent connection to *glass-fibers*

glass-crystal connection

Photonic Glass

Glass with 2nd-order nonlinearity

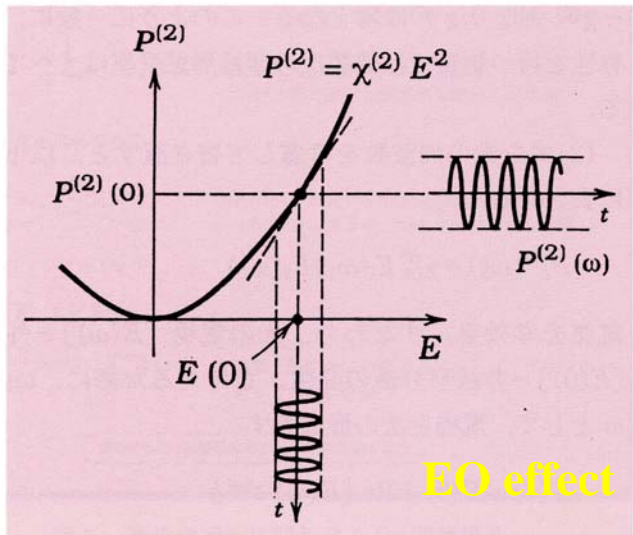
Alternative Description of Δn

EO effect (Pockels effect)

Electric field of angular frequency: $E(\omega)$

Applied electric field: $E(0)$

Nonlinear susceptibility: $\chi^{(2)}$



If $E(0) > E(\omega)$, 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 $E(0)$.

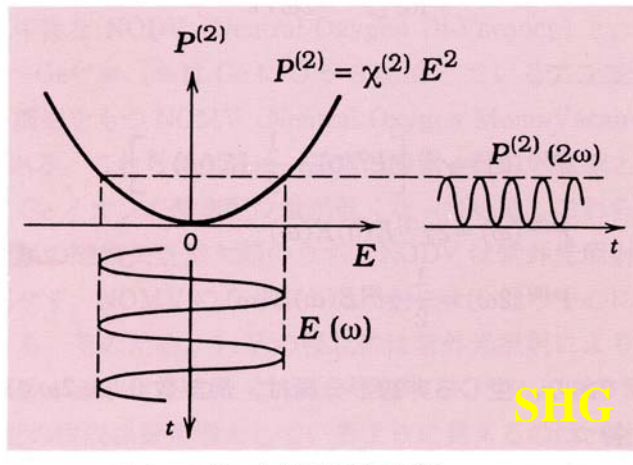
The corresponding incremental change of the refractive index is obtained by the relation $n^2 = 1 + \chi$, to obtain $2n\Delta n = \Delta \chi$, from which

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

$\Delta n = -rn^3E/2$ 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:

$$V = C_0 / n$$

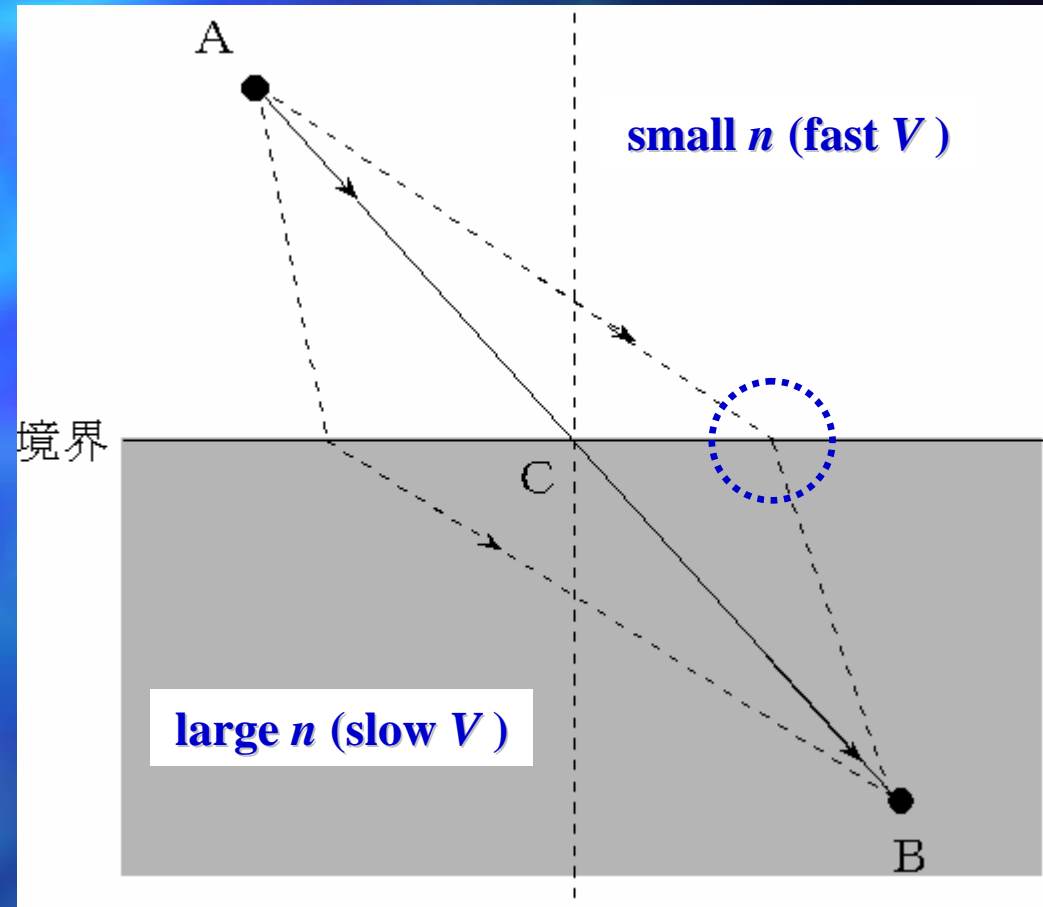
velocity in the medium: V

free space velocity: C_0

refractive index: n

large $n \rightarrow V$: slow

small $n \rightarrow V$: fast

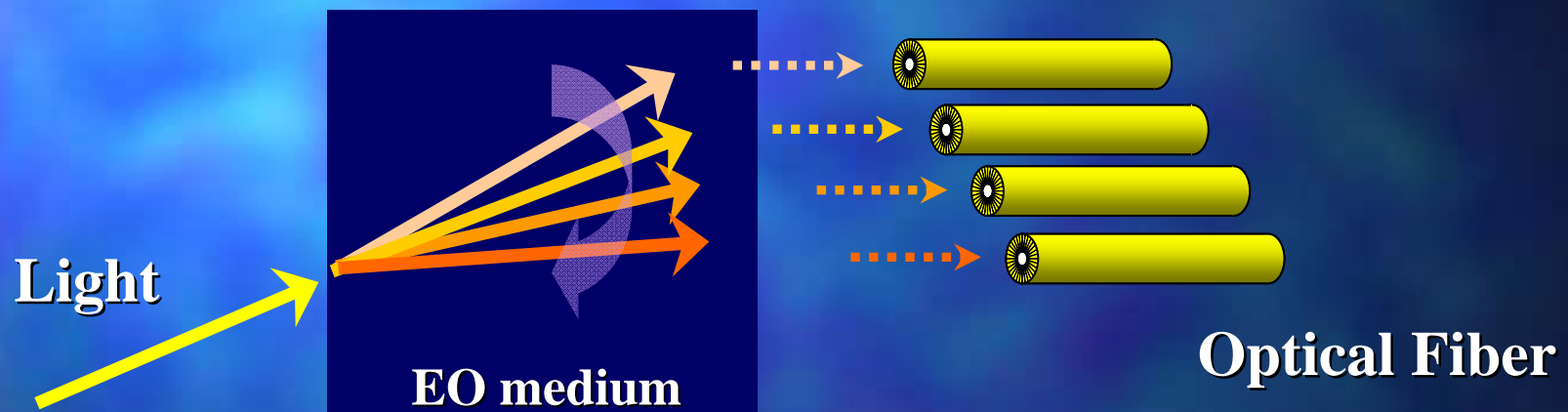


Light rays travel along the path of least time by refraction in this case (Snell' Law: $\sin \theta / \sin \phi = n_L / n_S$)

Controlling Light with EO Effect

Change of refractive index (Δn)

↳ Angle of refraction changed by E_{appl}

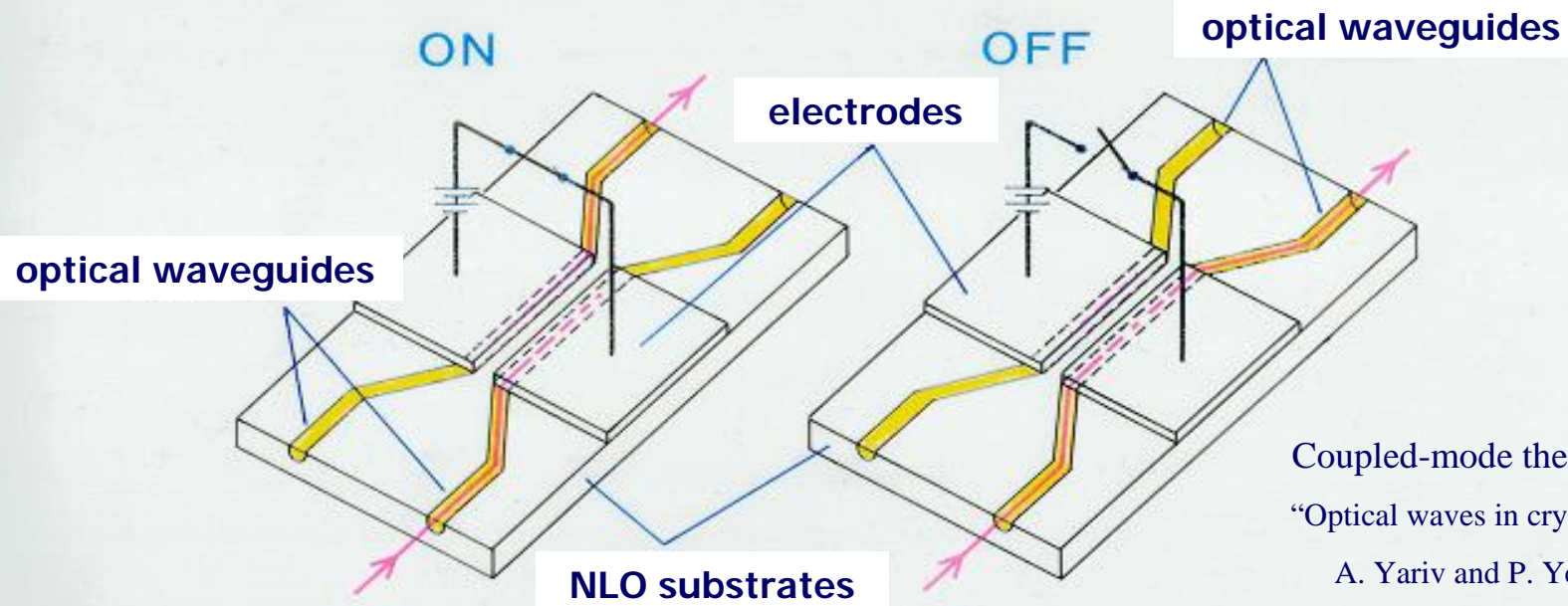


Controlling light with EO devices
through 2nd order optical nonlinearity

Electro-Optic Devices

Directional Coupler

2x2 Optical Switch



Coupled-mode theory :
"Optical waves in crystals"

A. Yariv and P. Yeh

Advantages of "Photonic Glass"

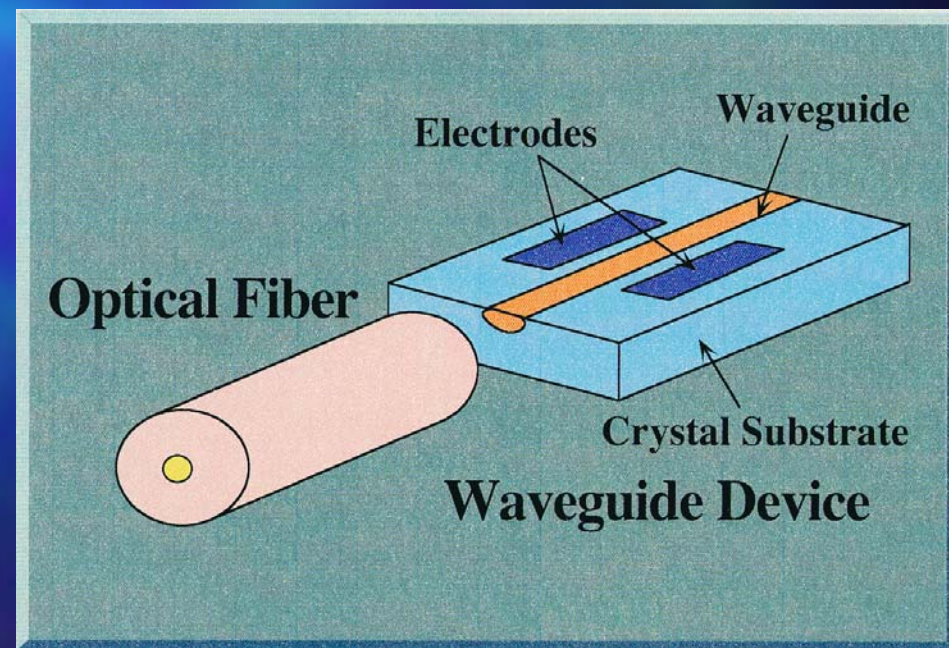
Advantages & Disadvantages

	Glasses	Inorganic materials	Organic materials
<u>2nd-Order NL</u>	X	○	⊙
Optical Loss	⊙	○	△
Transparent Range	⊙	△	△
Material Design	○	X	⊙
Connection	⊙	X	○
Shaping	⊙	△	△
Durability	⊙	△	△

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

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

***Second-Order Optical Nonlinearity**



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

2nd-order nonlinearity induced in glass

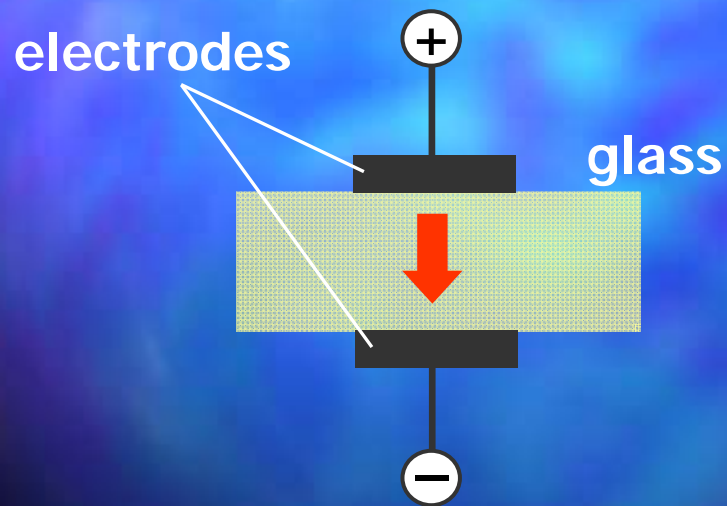
1. Poling with UV/heating
2. Crystallization

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

LiNbO₃ : ~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

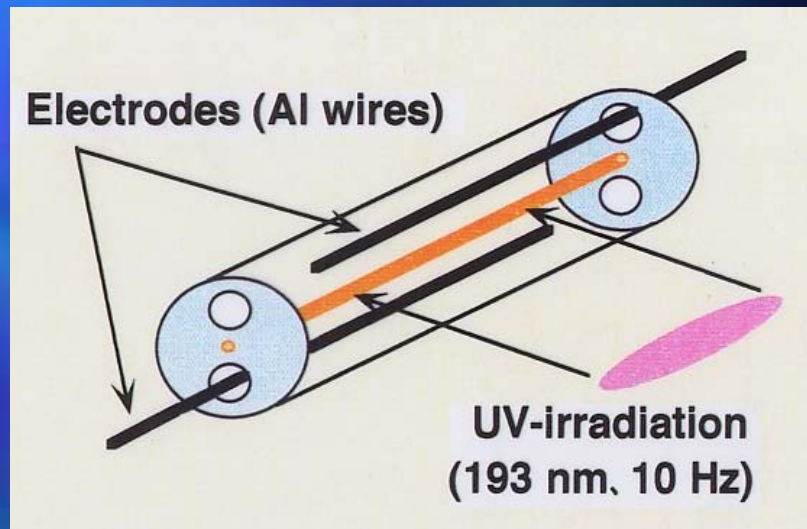
The Optical Fibre Technology Centre (OFTC)
University of Sydney, Australia

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
< 1 pm/V

UV-Poling in Ge:SiO₂ Fiber

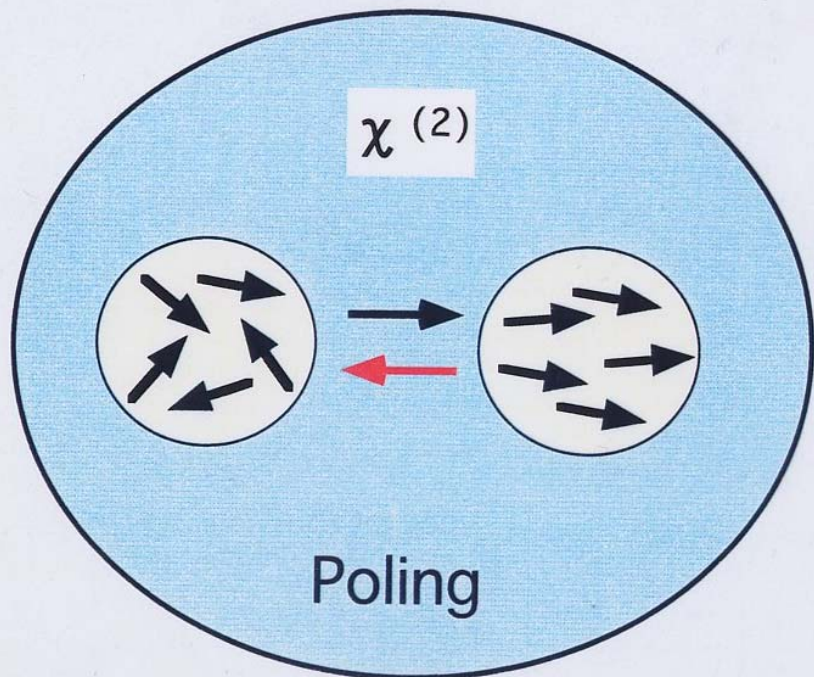


- Larger $\chi^{(2)}$: **~ 10 pm/V**
- Periodic structure: **$\chi^{(2)}$ gratings**
- Degradation** → *mechanism?*

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

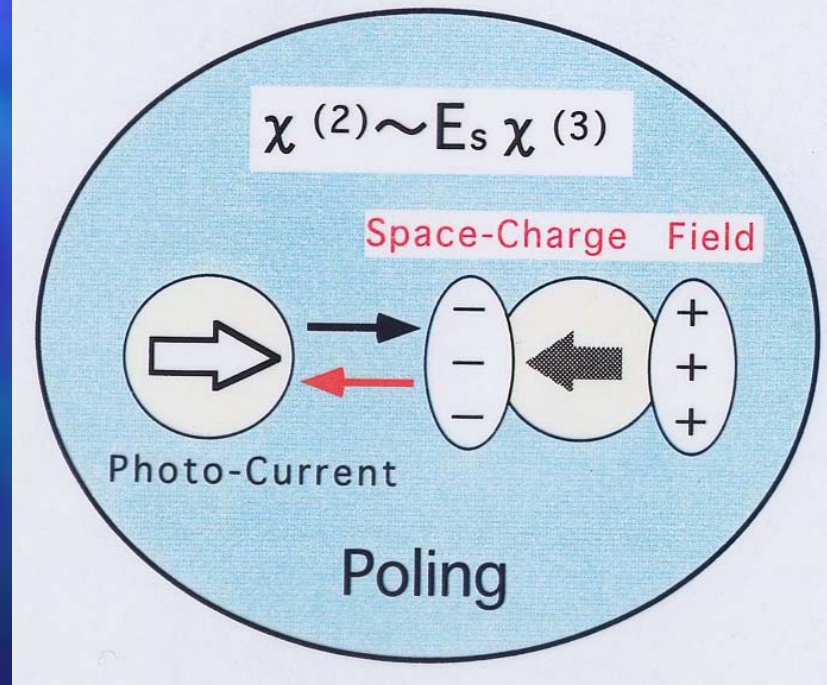
*Orientation of $\chi^{(2)}$
agents*

Dipole Orientation



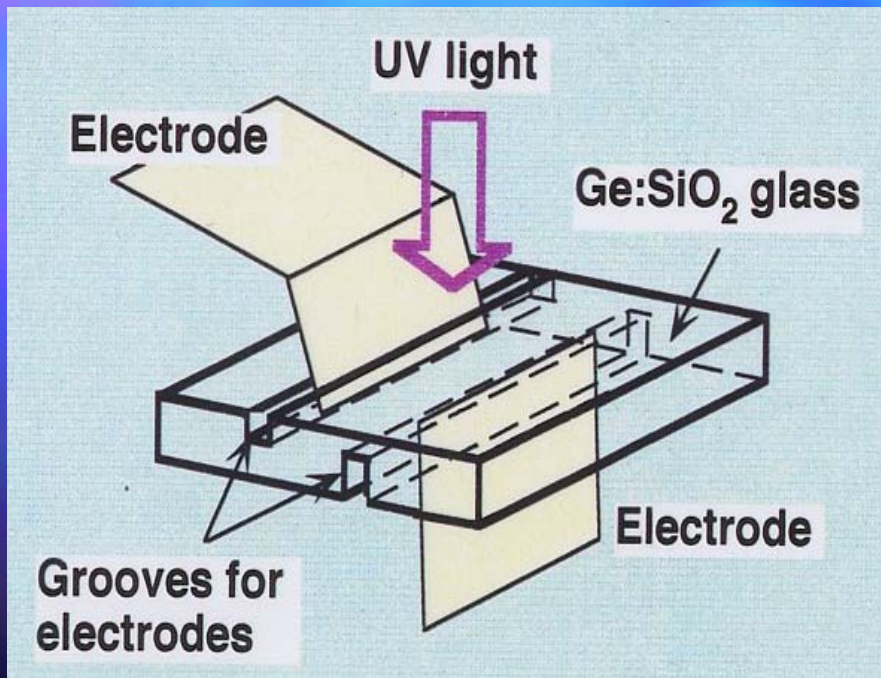
$\chi^{(2)}$ $EE \sim \chi^{(3)}$ EEE

Charge Separation



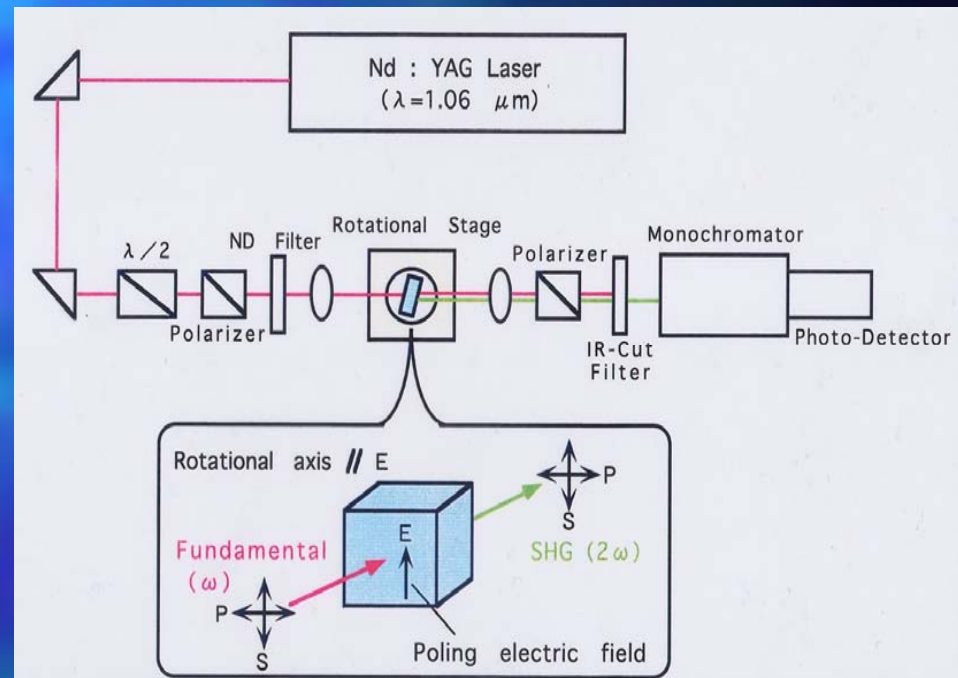
UV-Poling in Ge:SiO₂ Glass

UV-poling in bulk glass



- VAD preforms: 15GeO₂-85SiO₂
- E*-field: 0~3x10⁵ V/cm
- UV-laser: 193 nm

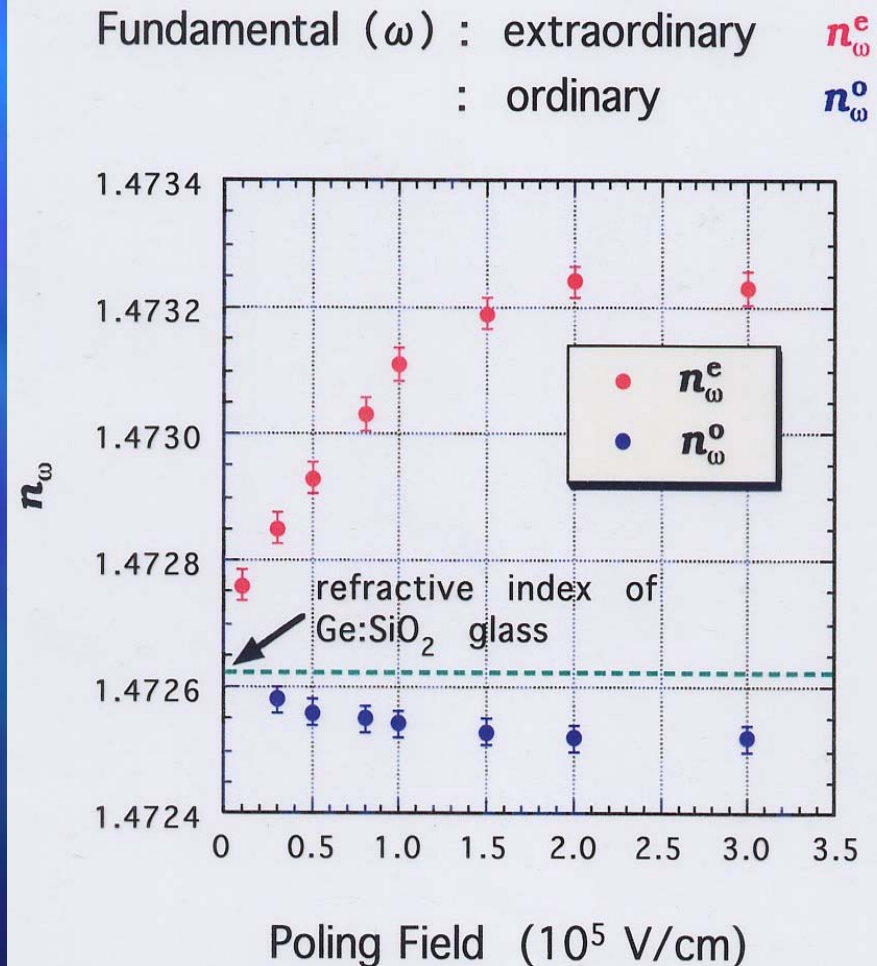
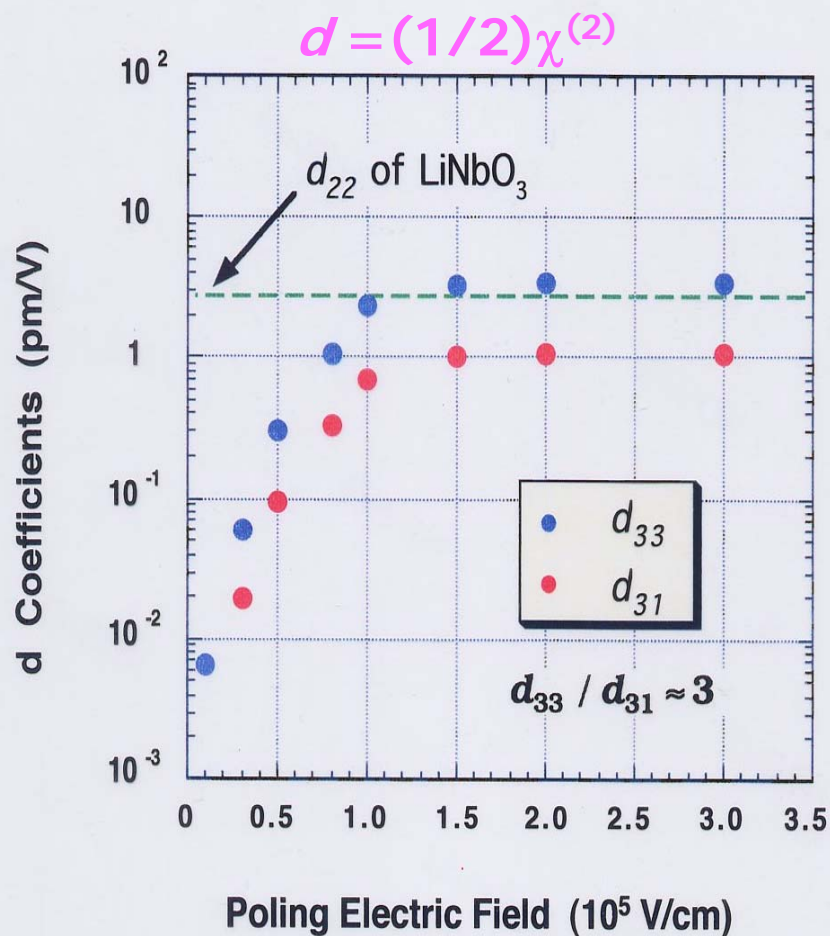
Maker-fringe SHG measurement



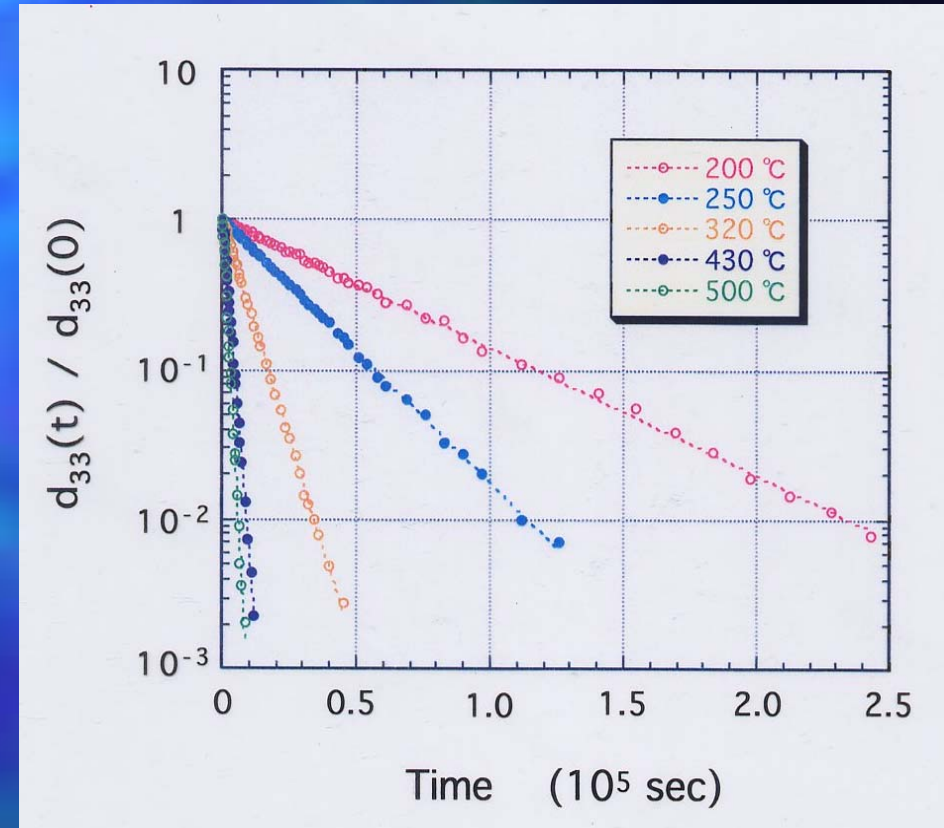
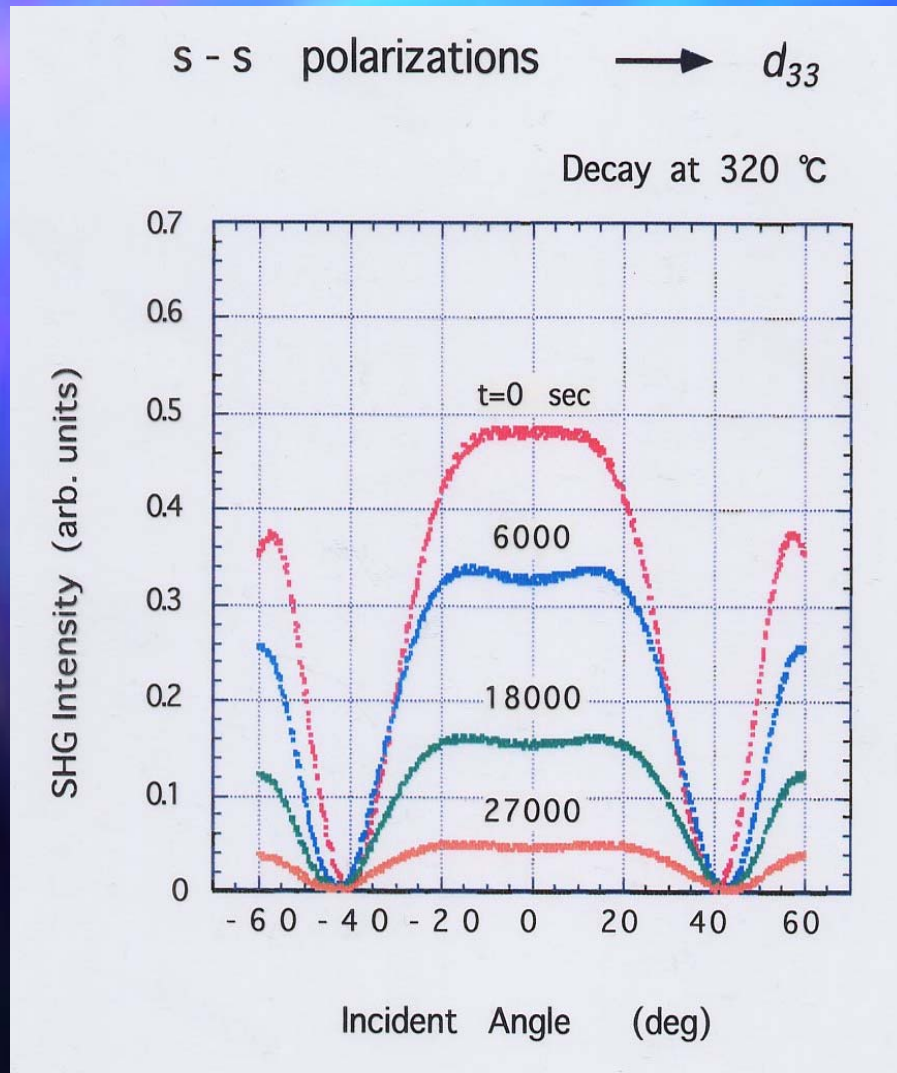
- Quantitative evaluation of SHG $d(\chi)$ coefficients
- Values of d_{33} , d_{31}
- Refractive index: n_e , n_o

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 SiO₂ Glass

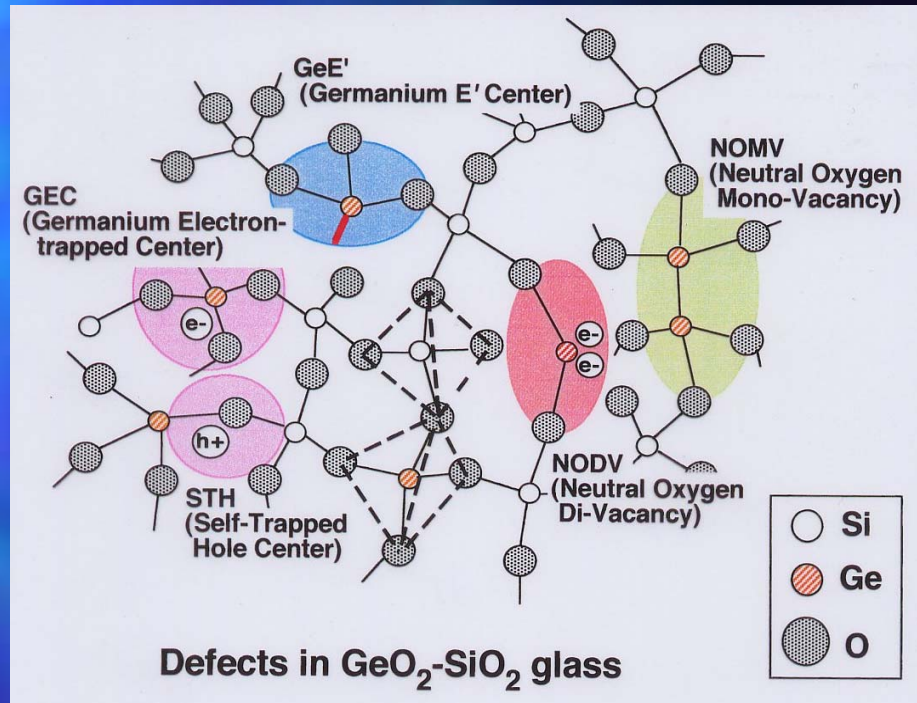
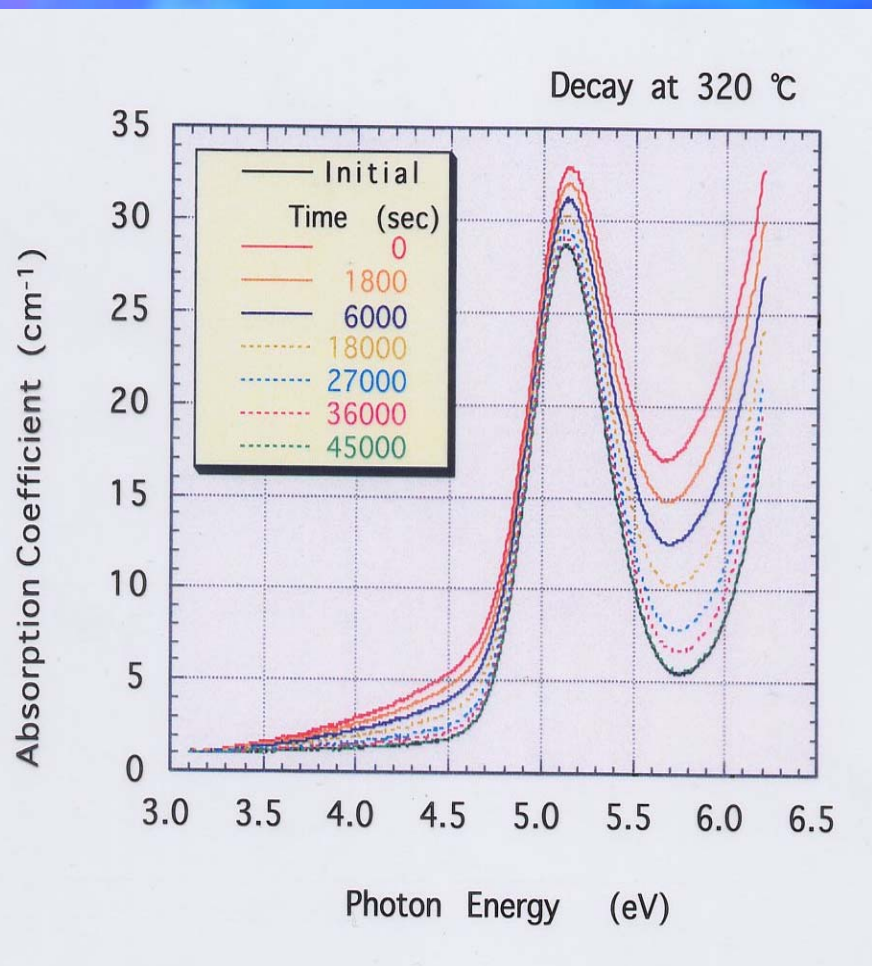
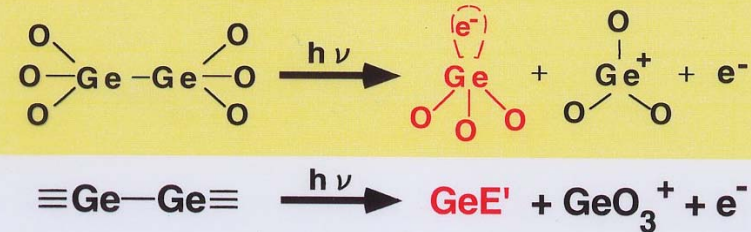
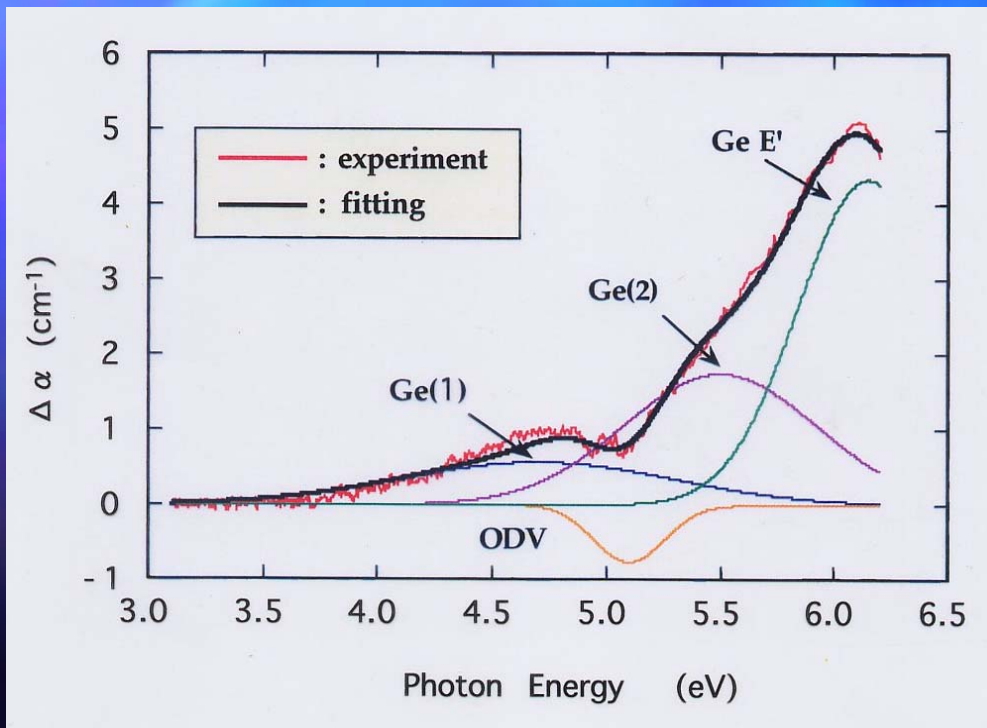


Photo-Chemical Reaction in GeO₂-SiO₂ glass

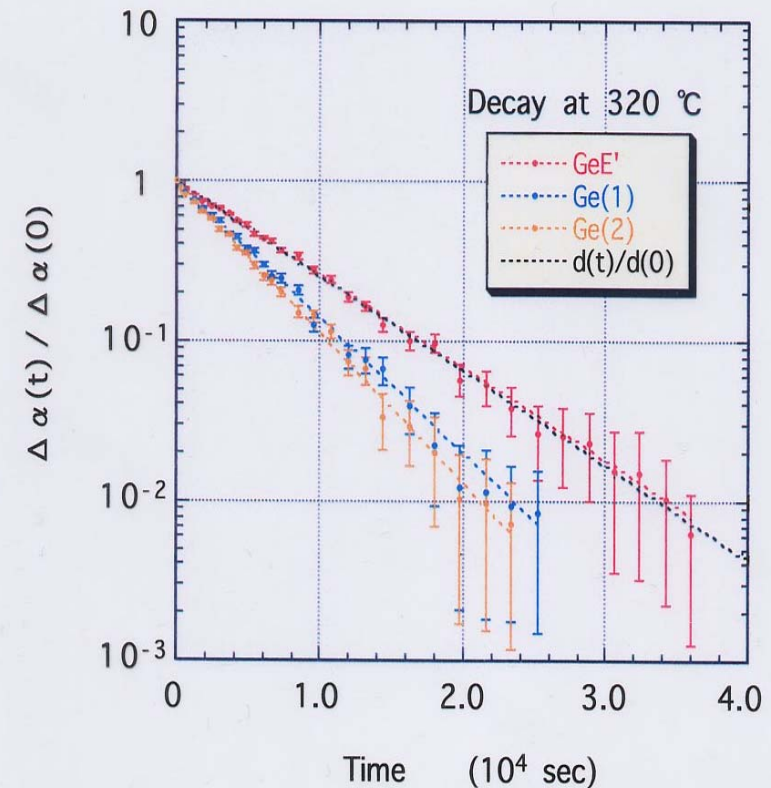


Quantitative Analysis of Decay (2)

Deconvolution of $\Delta\alpha$

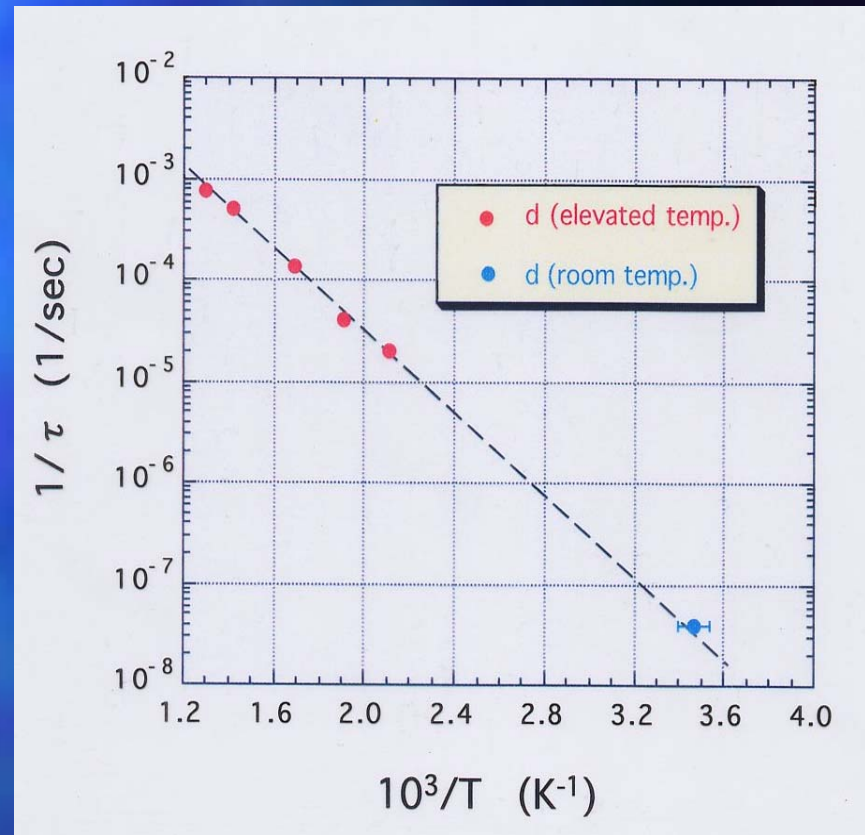
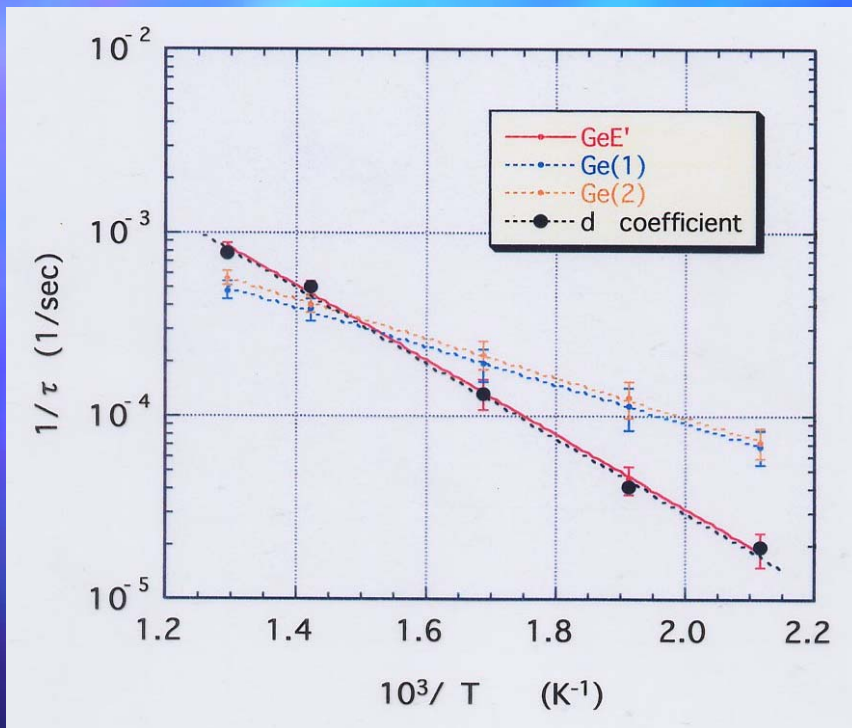


Decay of $\Delta\alpha$



$\chi^{(2)}$ 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

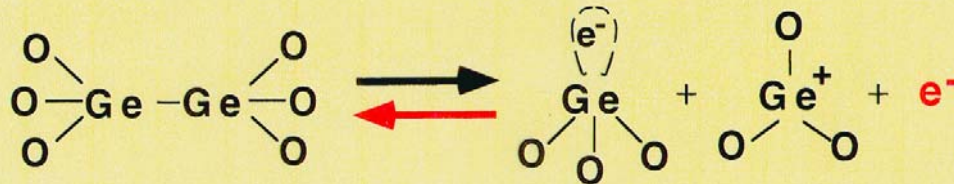
~280 days at RT

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

Comparison of activation energies

	Activation Energy (eV)
<i>Decay of d Coefficient</i>	
bulk (untreated)	• 0.41±0.05
bulk (heat treated)	0.38±0.05
<i>Dark Conductivity</i>	
bulk (untreated)	• 0.44±0.05
bulk (heat treated)	0.37±0.05
<i>Defects</i>	
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

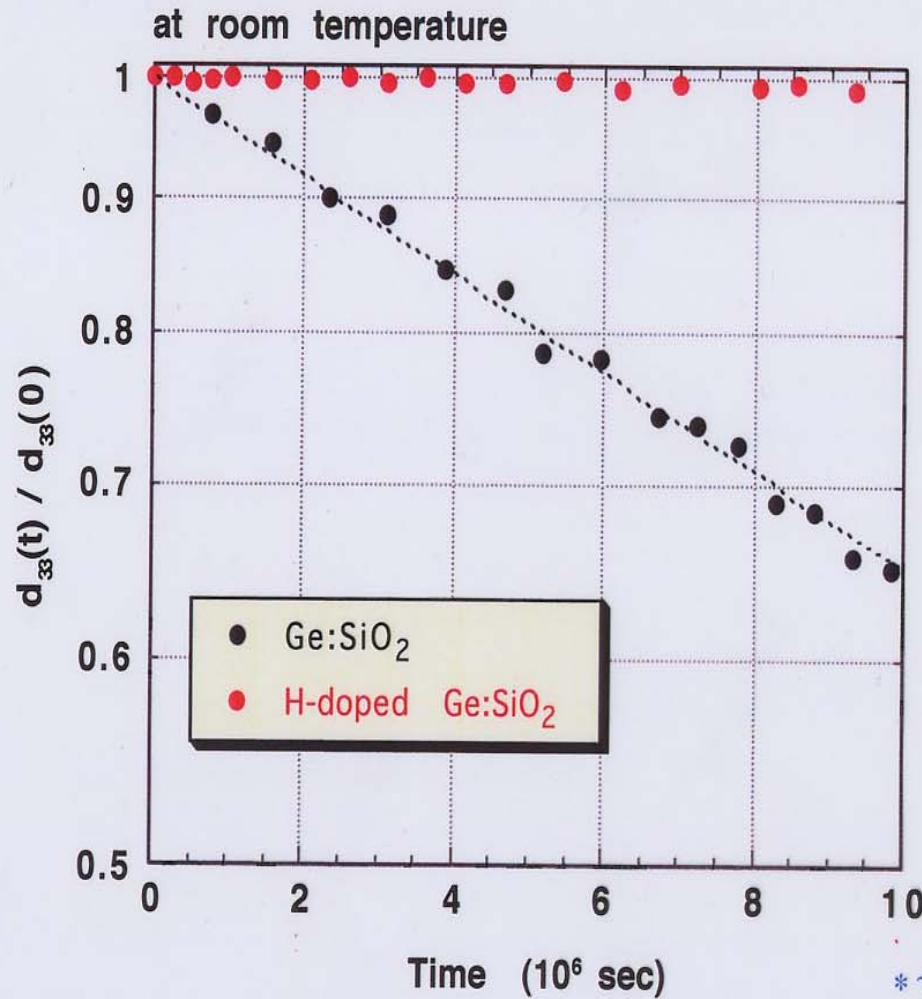
Introduction of electron scavengers?

For long-term stability



Hydrogen doping

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



$\tau \sim 18$ years*
 $d(0) = 4.5$ pm/V

$\tau \sim 280$ days*
 $d(0) = 3.4$ pm/V

application to
real field

For 20 years,
>90% performance

* τ : decay time constant
extrapolated from Arrhenius plot

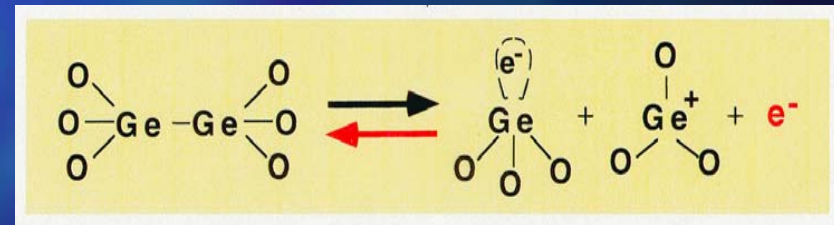
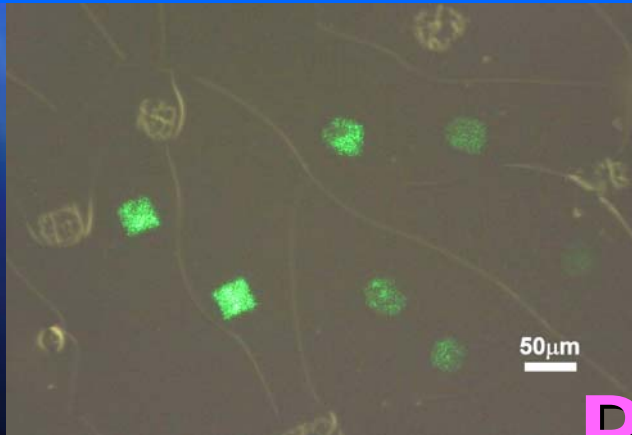
Origin and Decay of $\chi^{(2)}$ in UV-Poled Glass

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

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

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

E_{sc} : space-charge field caused by defect formation

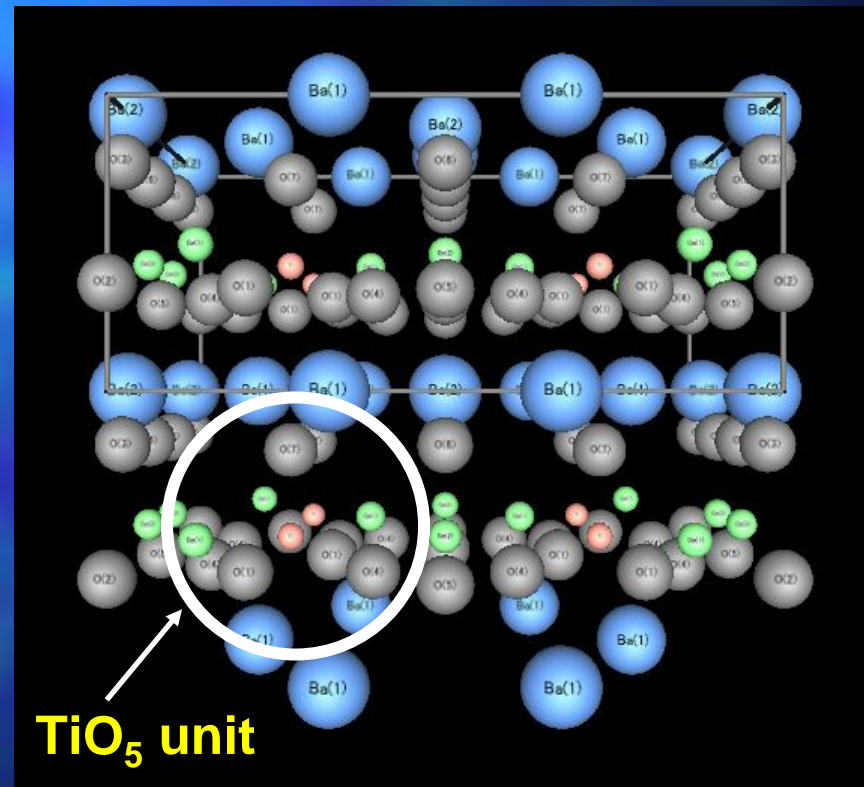
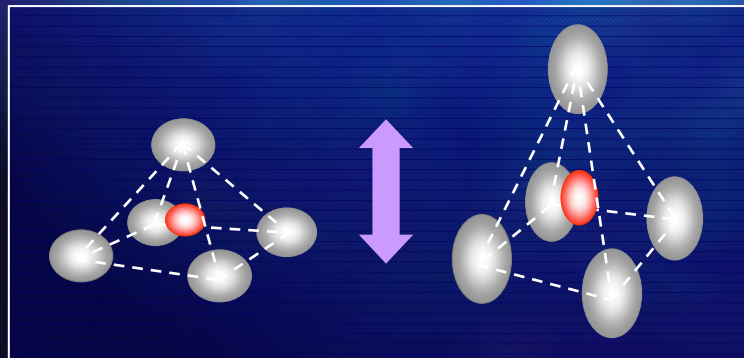
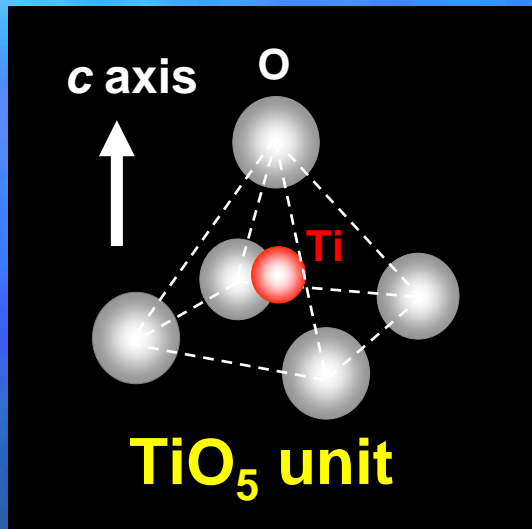


Permanent $\chi^{(2)}$?

Ba₂TiGe₂O₈ (BTG)

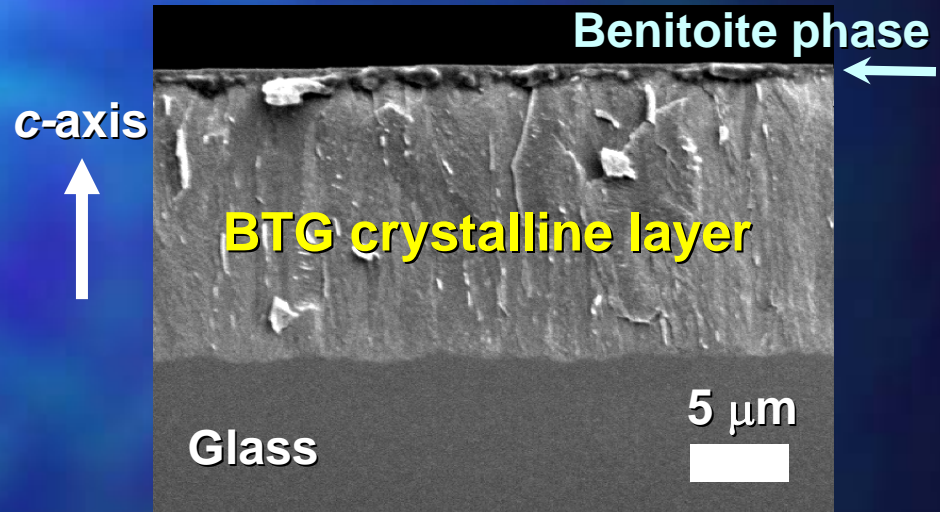
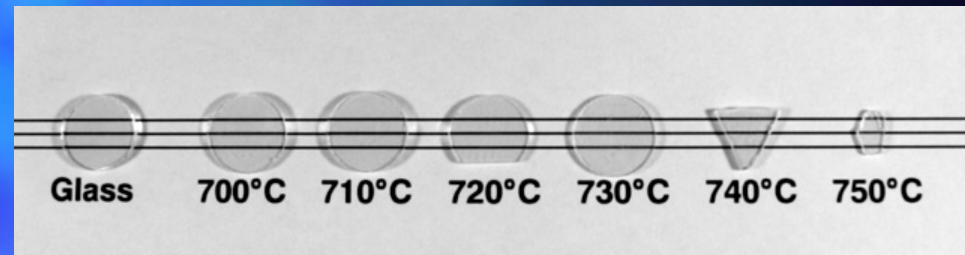
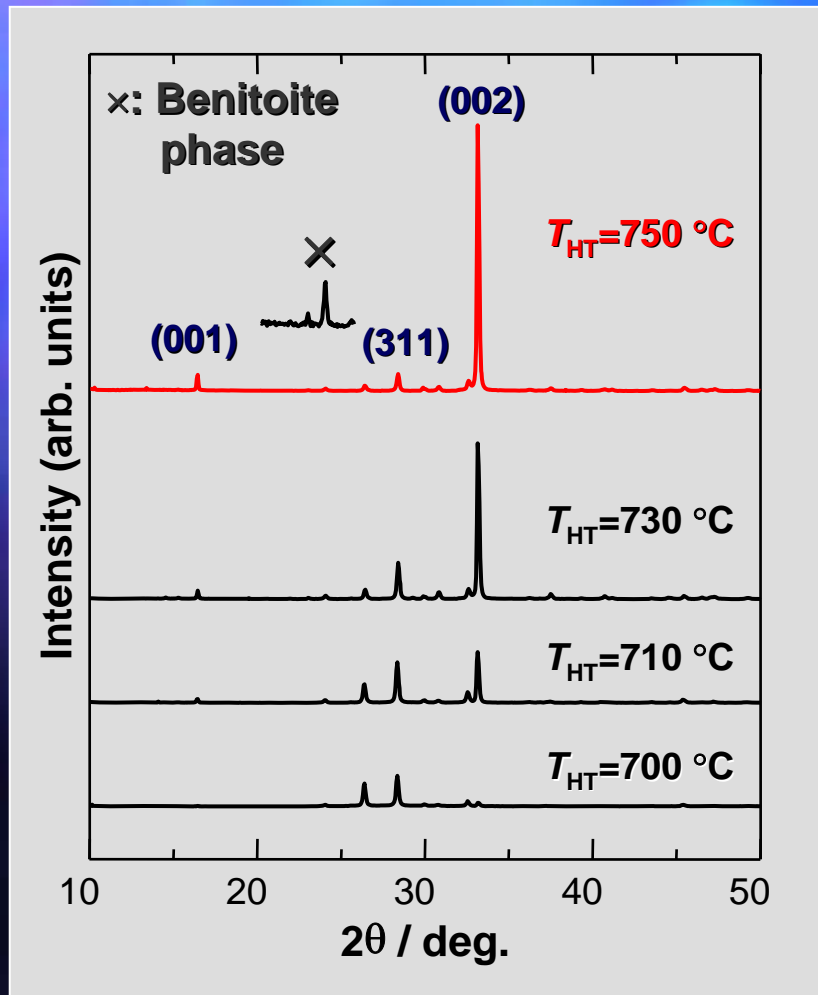
Fresnoite Crystalline Structure

Origin of P_s (spontaneous polarization)



Novel Crystallized Glass – BTG

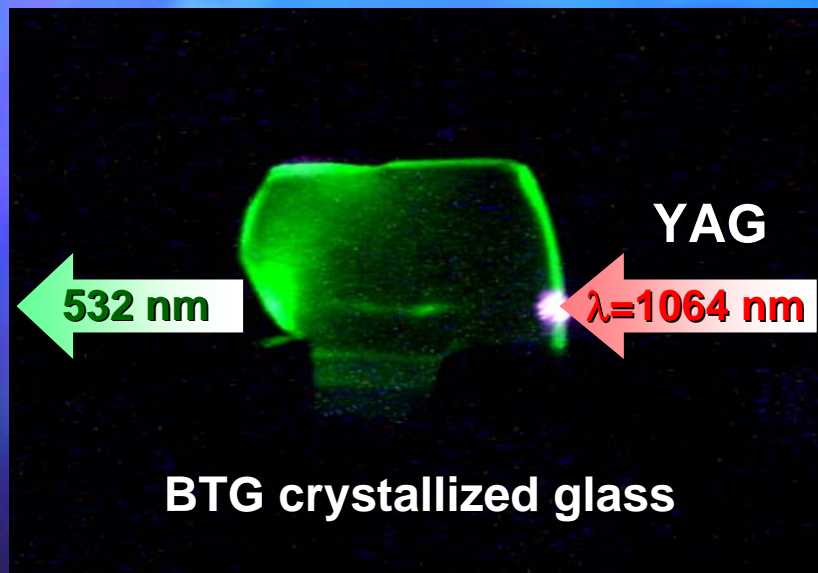
Surface Crystallization and Orientation



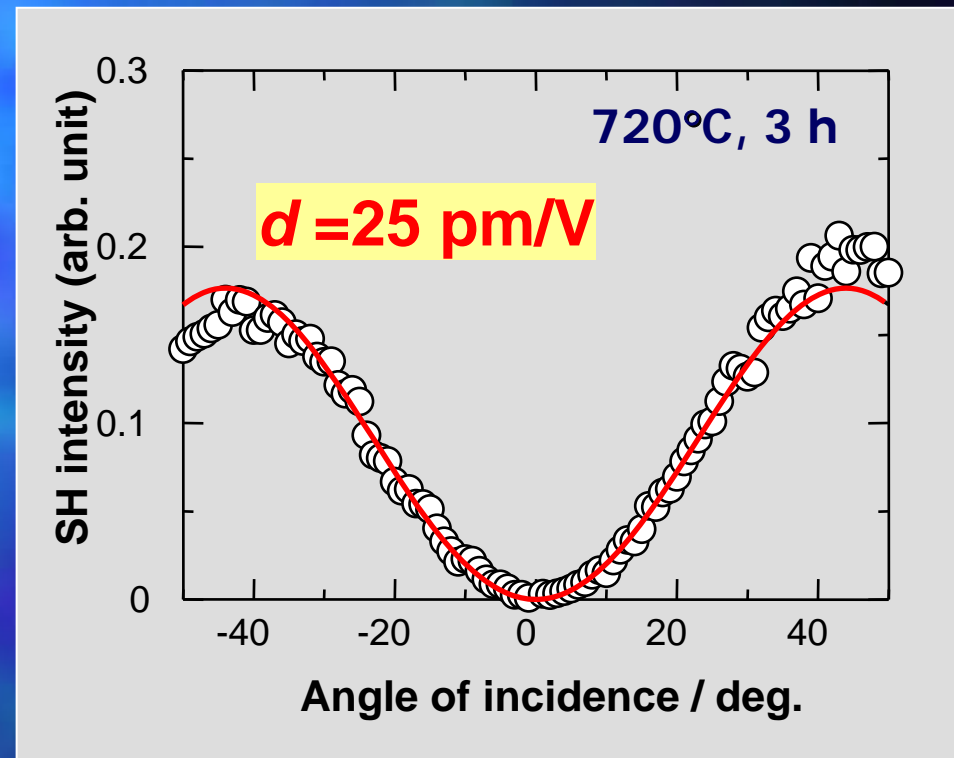
Stoichiometric composition

2nd-Order Nonlinearity in BTG

BTG55: 30BaO₂-15TiO₂-55GeO₂

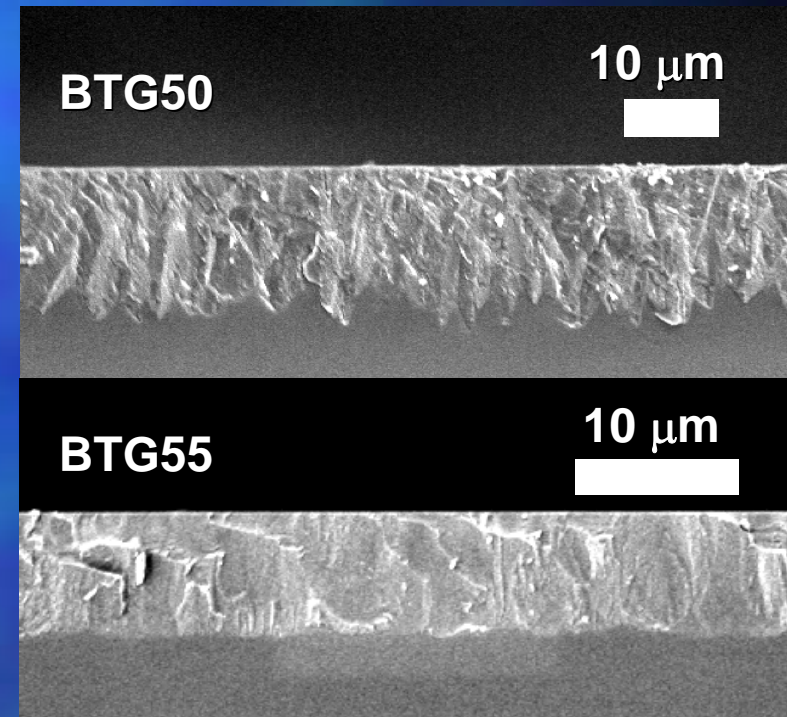
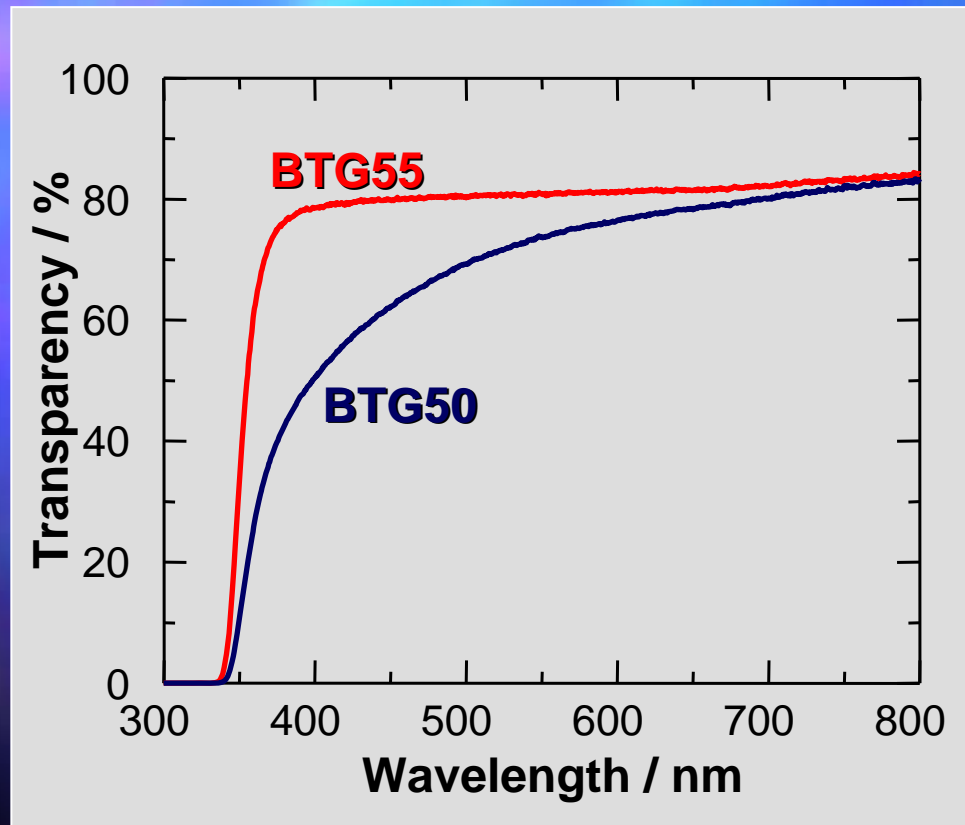


Appl. Phys. Lett., 81, 223(2002).



**Maker fringe measurement:
The largest *d*-value in glass ever reported**

Optical Absorption and Microstructure of BTG55 and BTG50



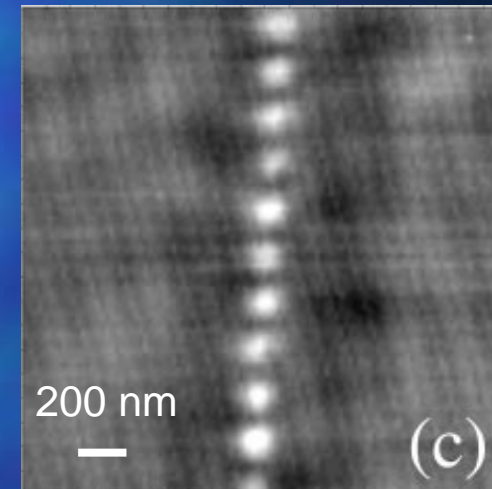
Crystalline layer of BTG55 is more dense and homogeneous than those of BTG50.

Plasmonics

Surface plasmon localized 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)

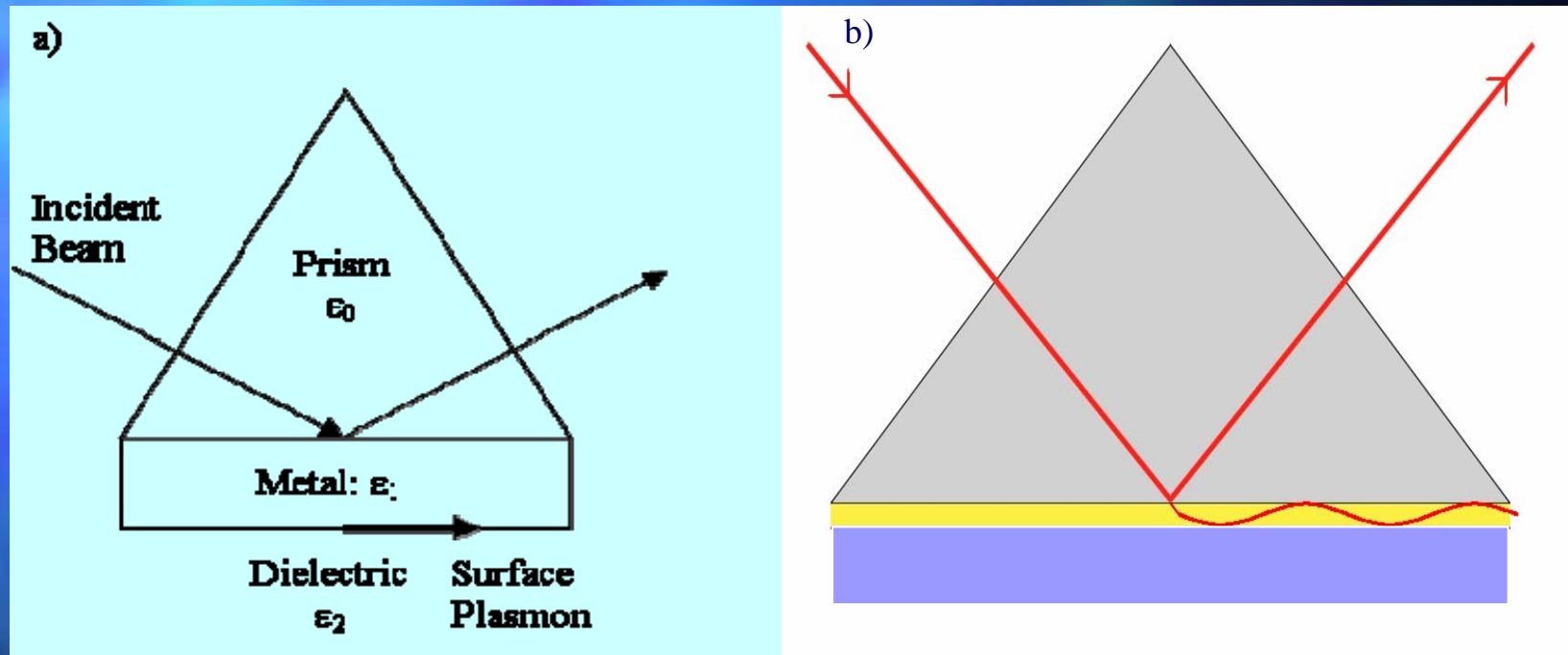


Optical intensity image of Au nano-particles ordering in glass substrate

J. of Microscopy, 202, (2001) 122

Surface Plasmon (SP)

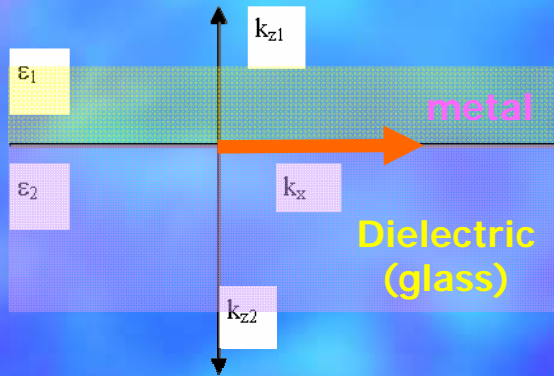
1. Excitation of SP by photon coupling



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.

Surface Plasmon (SP)

2. Dispersion relationship for SP



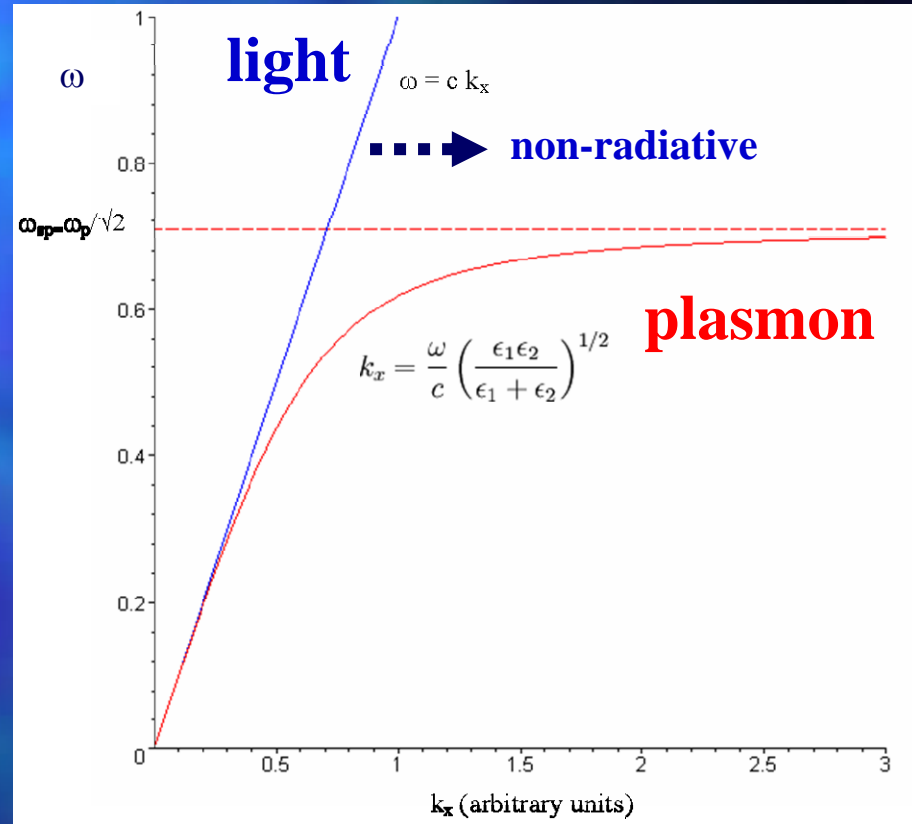
Wave number of SP: k_x
 Dielectric constants (relative): ϵ_1 and ϵ_2
 for metal and dielectric, respectively.

$$k_x = \frac{\omega}{c} \left(\frac{\epsilon_1 \epsilon_2}{\epsilon_1 + \epsilon_2} \right)^{1/2}$$

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

Since $\epsilon_1 < 0$ in metal, for the solution of k_x (plasmon),

$$\epsilon_1(\omega) < -\epsilon_2, \text{ below } \omega_{sp}$$



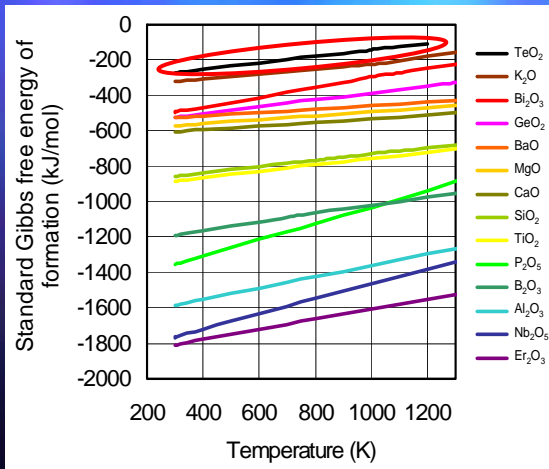
Dispersion curve for surface plasmons. At low k , the surface plasmon curve (red) approaches the photon curve (blue).

Laser-Induced Structure Ordering

Tellurite-based glasses

- Nano-crystallization by laser heating
- Selective crystallization of metal Te?
- Large nonlinearity: $d \sim 30d$ (LiNbO₃)

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



KNbO₃-TeO₂ glass



Periodic Structure with PM

XeCl excimer laser ($\lambda=308\text{nm}$)



Phase Mask (PM)



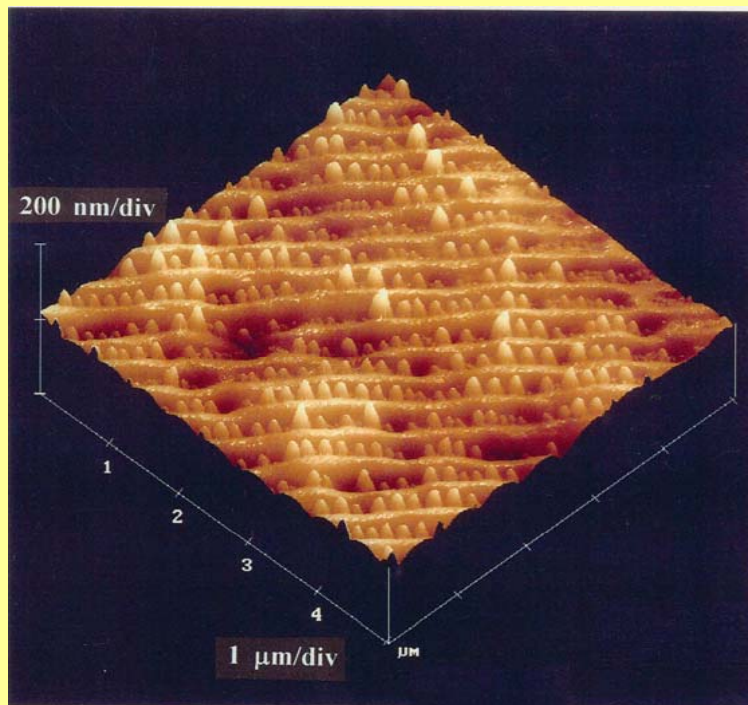
KNbO₃-TeO₂ glass

Photo-Induced Nano-Crystallization by UV-Laser Irradiations

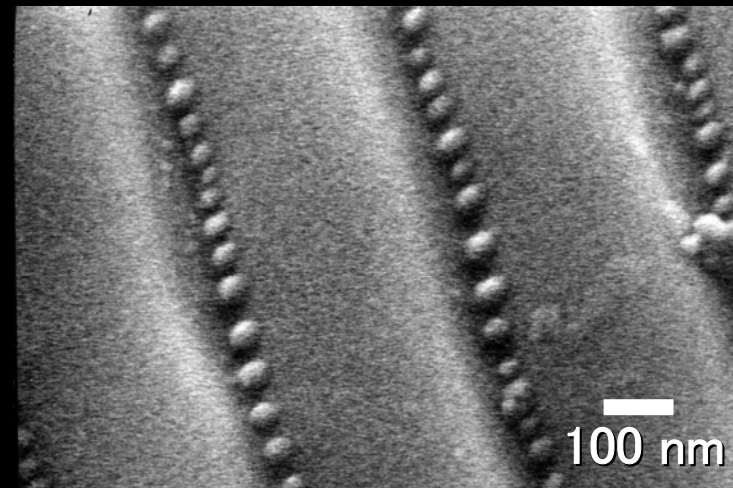
Periodic Structures of Nano-Particles 2

Structure Ordering in Glass

AFM image (enlarged)



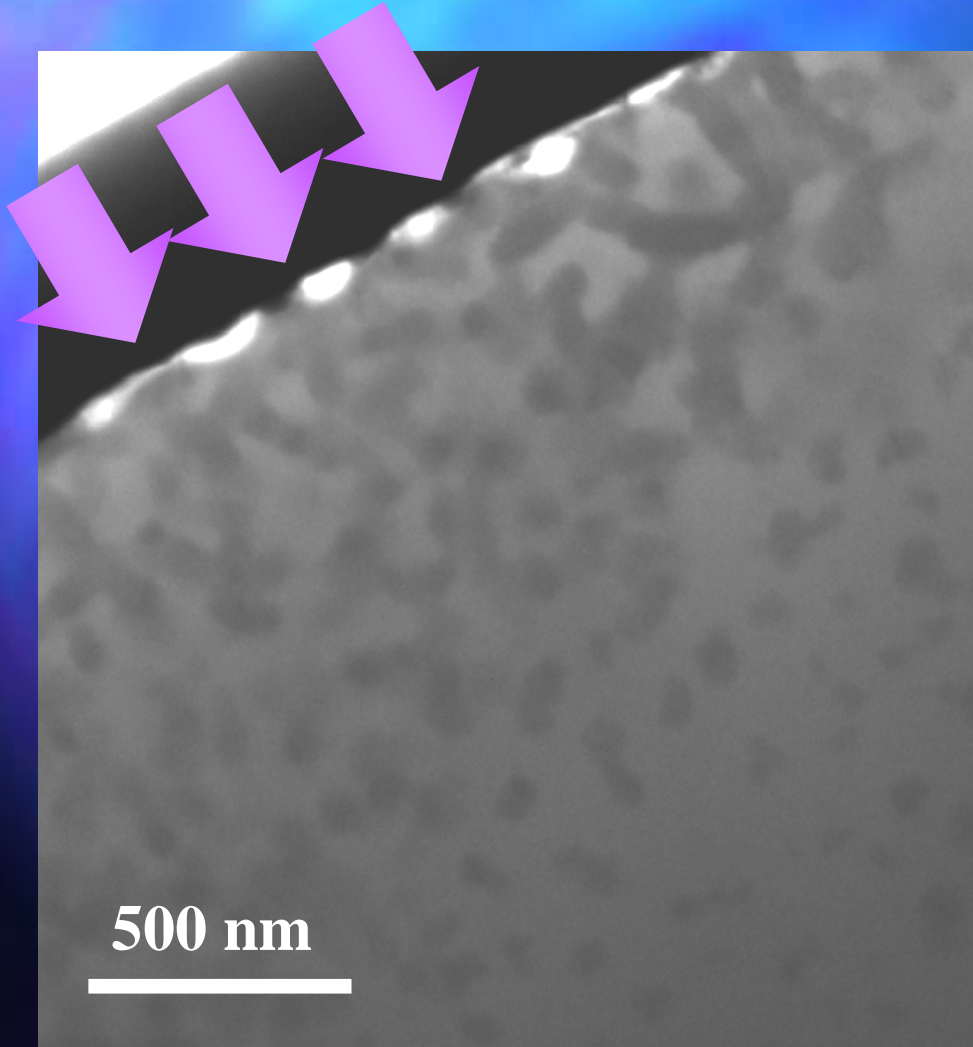
SEM image



ordered structure of
nano-particles

TEM Images of Surface Cross-Section

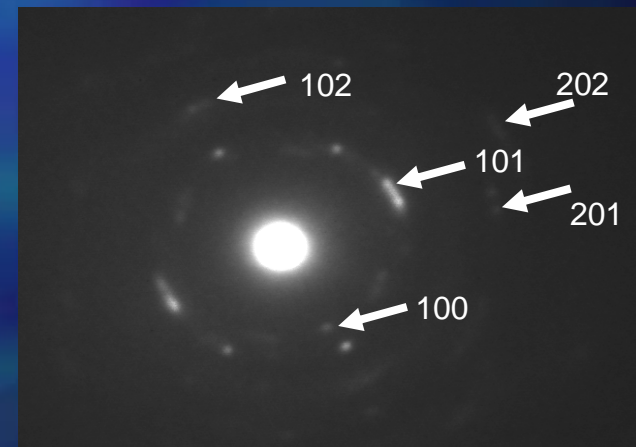
UV-Irradiation



- 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

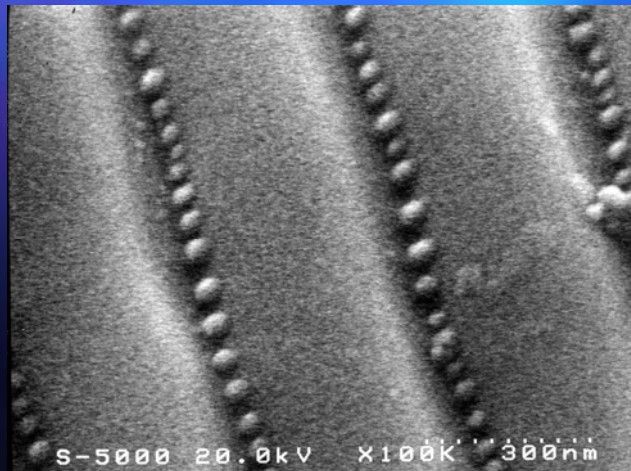
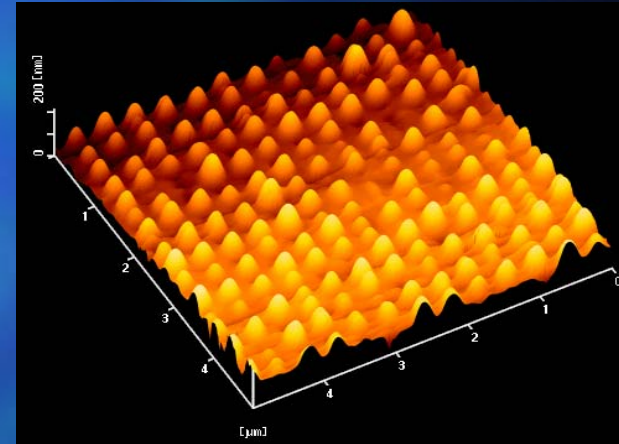
Plasmonic Glass



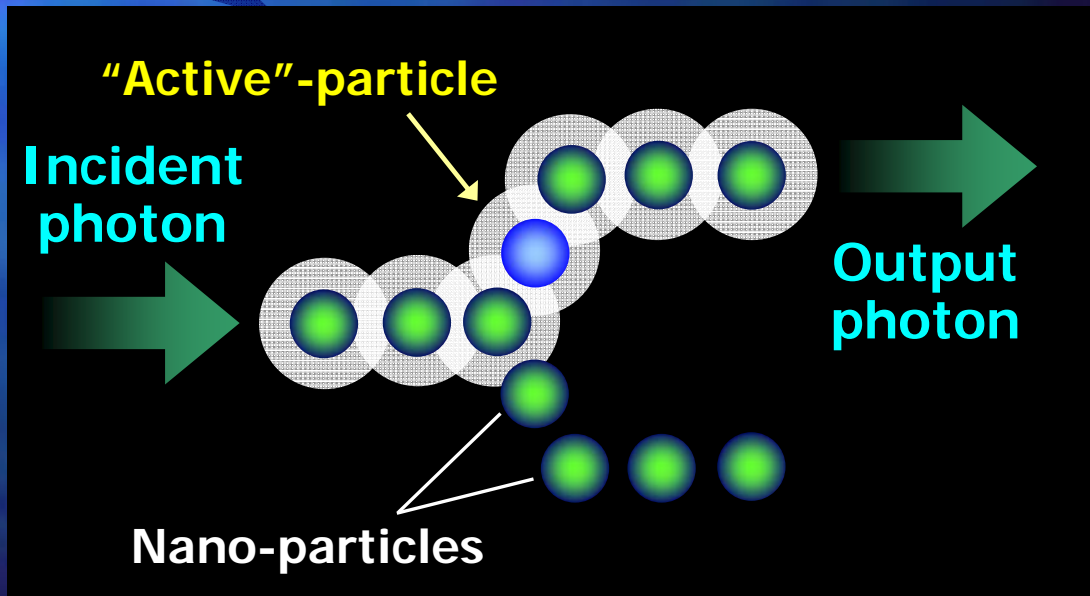
Plasmonic Glass for Nano-Circuit

Photo-Induced Nano-Particles Structure

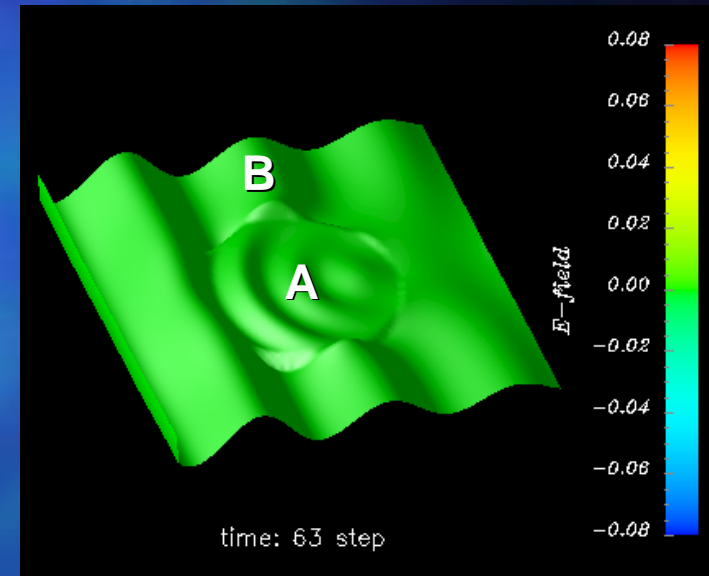
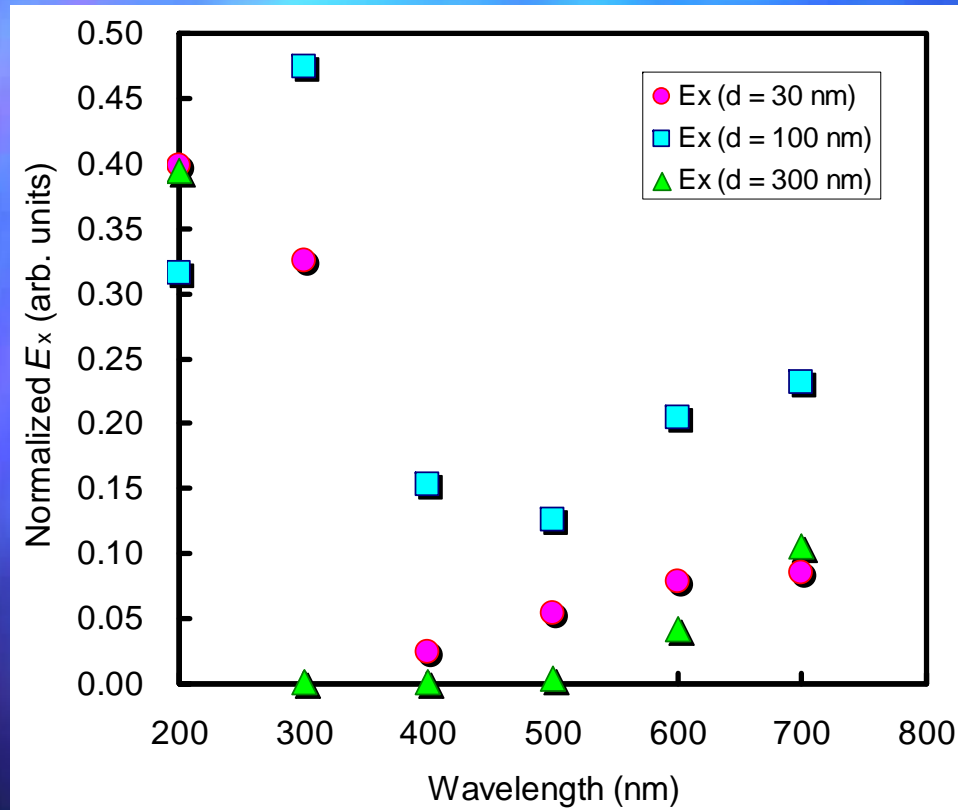
- Metal nano-particles on glass
- Physics for formation
- Design and control of particles
- **Nano-photonic circuits**



Ordered Nano-Particle Structure



Change of E-field Intensity (FDTD)



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

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 nanoparticle structures on glass surface

Plasmonic Glass

for Propagation/Localization of Light

