INTERNATIONAL ATOMIC ENERGY AGENCY
UNITED NATIONS EDUCATIONAL, SCIENTIFIC AND CULTURAL ORGANIZATION
INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS
I.C.T.P., P.O. BOX 586, 34100 TRIESTE, ITALY, CABLE: CENTRATOM TRIESTE

H4.SMR/845-9

Second Winter College on Optics

20 February - 10 March 1995

Photorefractive Effect and Applications Part II

K. Singh

Indian Institute of Technology
Department of Physics
New Delhi, India

SECOND WINTER COLLEGE ON OPTICS
ICTP, TRIESTE (ITALY); 20 FEB–10 MARCH 1995

PHOTOREFRACTIVE EFFECT, MATERIALS AND APPLICATIONS

KEHAR SINGH

PHOTONICS GROUP, PHYSICS DEPARTMENT
INDIAN INSTITUTE OF TECHNOLOGY
HAUZ KHAS, DELHI, NEW–DELHI 110016
INDIA

FAX: 91–11–6862037
TELEX: 31–73087 IITD IN
31–61460 IITD IN
e–mail: kehars @ physics.iitd.ernet.in

PHOTONIC/OPTICAL ARTIFICIAL INTELLIGENCE

[I THINK THEREFORE I AM - DESCARTES]

* LIVING IN INFORMATION AGE
* KNOWLEDGE IS POWER
* NATIONS AIM AT ESTABLISHING KNOWLEDGE INDUSTRY
* SALABLE COMMODITY (LIKE OIL,FOOD,...)
* COMPUTER - AMPLIFIER OF THIS POWER
  (OPTICAL COMPUTER - SUPER AMPLIFIER)

INFORMATION STORAGE AND INFORMATION PROCESSING

* OPTICS MAKING RAPID INROADS INTO AREAS HITHERTO DOMINATED BY ELECTRONICS
* ASTONISHING IN POTENTIAL TO RESHAPE ELECTRONICS-CENTERED TECHNOLOGIES
* BASIC PERCEPTION + MOTOR SKILLS
  EVEN LOWER ANIMALS POSSESS PHENOMENAL CAPABILITIES COMPARED TO COMPUTERS
* OPTICS PROVIDES: ADAPTIVE, MASSIVELY PARALLEL DENSLY CONNECTED ARCHITECTURE, CONNECTIONS -> MODIFIABLE

- OPTICS MAKING INROADS INTO APPLICATION AREAS HITHERTO DOMINATED BY ELECTRONICS

  (BANDWIDTH, ELECTR. INTERF.)

- MATERIAL LIMITATIONS SLOWING DOWN DEVELOPMENTS.

  (DEVICE QUALITY MATERIALS
  LARGE NONLINEAR RESPONSE)

  ACTIVE DEVICES.

ENGINEERING OF DESIRABLE OPTICAL PROPERTIES

TWO-APPROACHES

- SUBPICOSECOND SPEEDS (LITTLE/NO COMPETITION FROM ELECTRONICS)
- SLOWER DEVICES

  HIGH DENSITY INTERCONNECTIONS

APPLICATIONS

PHOTONICS (LIGHT ELECTRONICS)

- ARTIFICIAL INTELLIGENCE
- MACHINE/ROBOTIC VISION
  - AUTOMATIC PATTERN RECOGNITION
  - AUTOMATED PRODUCT INSPECTION
  - DATA & KNOWLEDGE BASE SYSTEM
  - OPTICAL COMPUTING
  - OPT. IMPLEMENTATION OF NEURAL NETS
- INTERFEROMETRIC TESTING
- PHOTOLITHOGRAPHY
- 3-D PICTORIAL INFORMATION TRANSMISSION (M.M. FIBERS)
- PHASE CONJUGATE LASERS
- INCOHERENT-TO-COHERENT CONVERTER
- OPTICAL LOGIC
- OPTICAL COMPUTING
- ASSOCIATIVE MEMORIES
- NOVELTY TRACKING FILTERS
- PHASE LOCKING OF LASERS

PATTERN RECOGNITION APPLICATIONS

- IMAGE PROCESSING, SEGMENTATION, AND ANALYSIS
- COMPUTER VISION
- SEISMIC ANALYSIS
- RADAR SIGNAL CLASSIFICATION / ANALYSIS
- FACE RECOGNITION
- SPEECH RECOGNITION / UNDERSTANDING
- FINGER PRINT IDENTIFICATION
- CHARACTER (LETTER OR NUMBER) RECOGNITION
- HANDWRITING ANALYSIS (NOTEPAD COMPUTERS)
- ELECTROCARDIOGRAPHY SIGNAL ANALYSIS
- MEDICAL DIAGNOSIS

NONLINEAR PHOTOREFRACTIVES

WIDER CLASS OF
MATERIALS

↓

NONLINEAR OPTICAL MATERIALS

ATOMIC VAPOURS
DYES
POLYMERS
LIQUID CRYSTALS
PLASMAS
QUANTUM WELLS
SUPERLATTICES

NON-LINEAR OPC

VARIOUS TECHNIQUES

- PHOTON ECHOES
- STIMULATED BRILLIOUN SCATTERING (SBS)
- SRS
- SQUEEZED STATES

FOOLHARDY TO EVEN THINK OF SUMMARIZING THE SUBJECT. TALL ORDER

SKETCHY TUTORIAL REVIEW: QUALITATIVE

OBJECTIVE : MERELY TO AROUSE CURIOSITY
GENERATE SOME MORE INTEREST

A REAL-ESTATE BROKER NAMED WHYST HAD A PHASE CONJUGATE HOUSE ON HER LIST THE CEILING WAS THE FLOOR AND THE WALL THE DOOR WHICH MADE HER EXCEEDINGLY PISST

PHASE CONJUGATION

STATIC (CONV. HOLOGR) → e.g. Ag HALIDES

DYNAMIC → NONLINEAR

LATE 60's : $LiNbO_3$ (1966)

OPTICAL DAMAGE : REF. INDEX CHANGE
NUISANCE
(IRREVERSIBLE)

PHOTOREFRACTIVE EFFECT :

LITERAL MEANING : CHANGE IN R.I.
DUE TO LIGHT

CONVENTION/USAGE : REVERSIBLE CHANGE
IN REF. INDEX

ADVANCES IN MATERIAL SCIENCE

NONLINEAR EFFECTS AT LOW POWER LEVELS

(NONLIN. OPT. OF EARLY 60's $\sim 10^7 - 10^9$ W/cm^2)

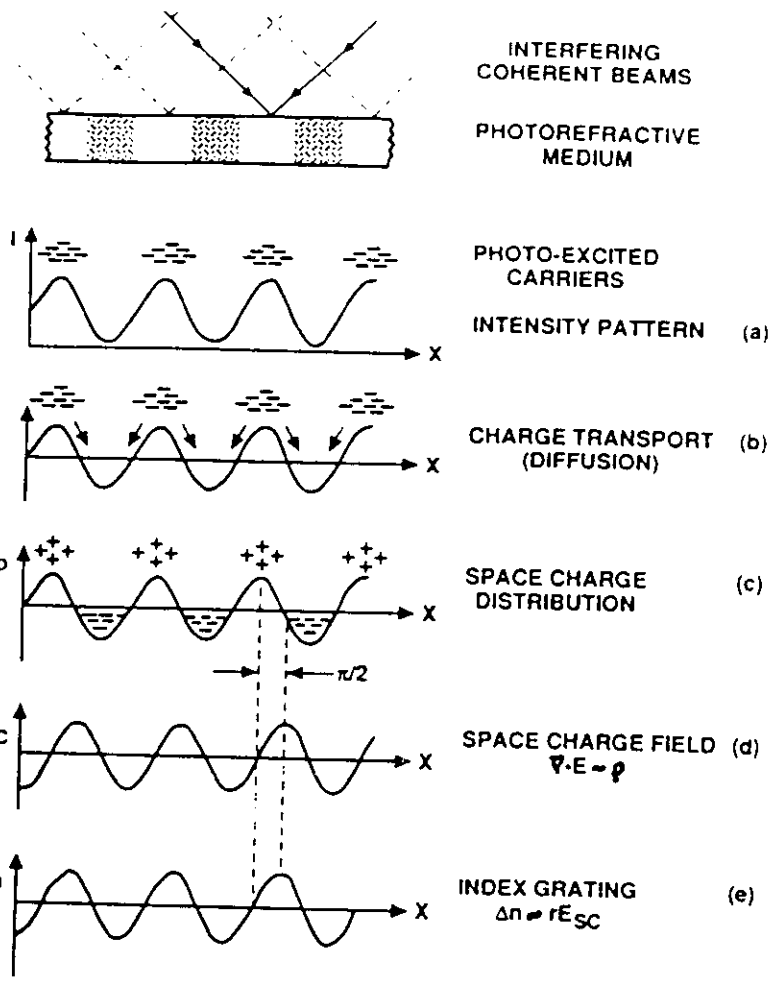


Figure The formation of photorefractive index gratings: (a) periodic light intensity pattern and photoexcited charge carriers; (b) charge transport by diffusion; (c) static charge distribution; (d) $\pi/2$ phase-shifted space-charge field distribution; (e) photorefractive index grating.

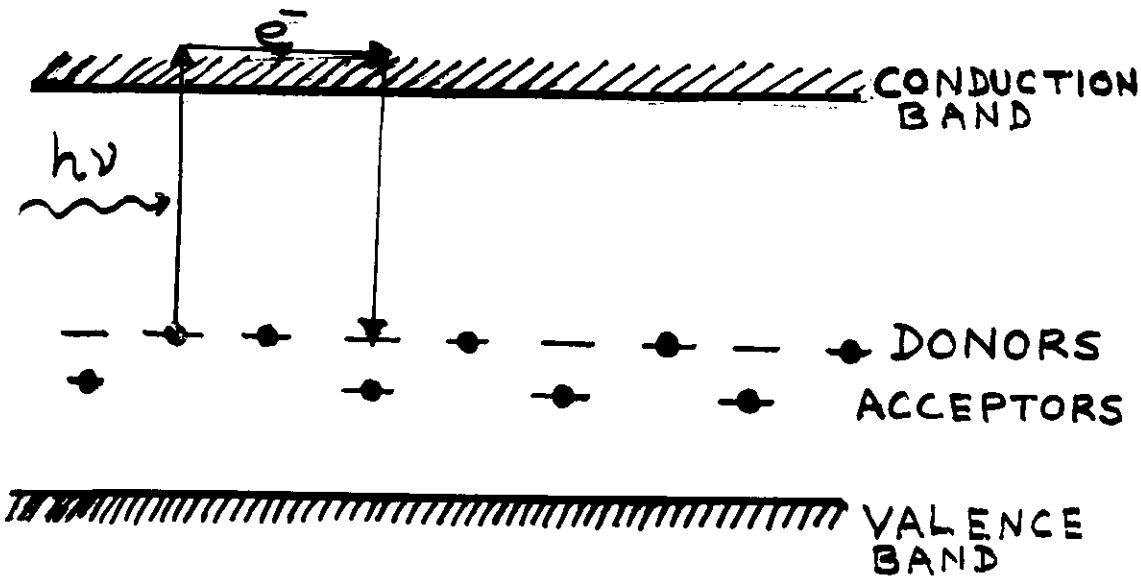
IN SUMMARY

P.R EFFECT BASICALLY CONSISTS OF FIVE FUNDAMENTAL PROCESSES OCCURING IN ELECTRO-OPTICAL CRYSTALS:

- PHOTOIONIZATION OF IMPURITIES AND GENERATION OF CHARGE CARRIERS
- TRANSPORT OF THESE CHARGE CARRIERS
- TRAPPING OF CHARGE CARRIERS AND FORMATION OF SPACE-CHARGE DENSITY
- FORMATION OF PHOTOINDUCED SPACE-CHARGE ELECTRIC FIELD
- FORMATION OF INDEX GRATING VIA LINEAR ELECTRO-OPTIC EFFECT (POCKEL'S EFFECT)
- UNIFORM ILLUMINATION ERASES SPACE-CHARGE

E.O. EFFECT:

APPLICATION OF ELECTRIC FIELD RESULTS IN A CHANGE IN THE DIELECTRIC PERMITTIVITY TENSOR ϵ ; OR EQUIVALENTLY A CHANGE IN BOTH THE DIMENSION AND ORIENTATION OF THE INDEX ELLIPSOID



SIMPLE MODEL : P.R. EFFECT

FAIRLY OFTEN ENCOUNTERED CASE

↓

DONORS AND TRAPS ARE IMPURITIES OF IONS OF ONE AND THE SAME TYPE OF ATOM BUT IN DIFFERENT VALENCE STATES

EXAMPLE :

Fe^{2+} AND Fe^{3+} IONS (IN OXIDE CRYSTALS)

EITHER IMPURITY OR DOPING IN $LiNbO_3$ AND $KNbO_3$, Fe IONS SUBSTITUTE FOR Nb^{5+}, THE LOCAL ELECTRONEUTRALITY BEING PRESERVED BY CREATION OF AN OXYGEN VACANCY NEAR Fe^{2+},

i.e AN Fe^{2+}-V_O CENTER IS FORMED.

TYPICAL ENERGIES FOR PHOTOEXCITATION OF Fe^{2+} IONS IN $LiNbO_3$ & $KNbO_3$ ARE 3.1 – 3.2 eV.

$\ominus \longleftrightarrow \oplus$

DONOR TRAP

(e.g. Fe^{2+}) (e.g. Fe^{3+})

GENERATION PROCESS

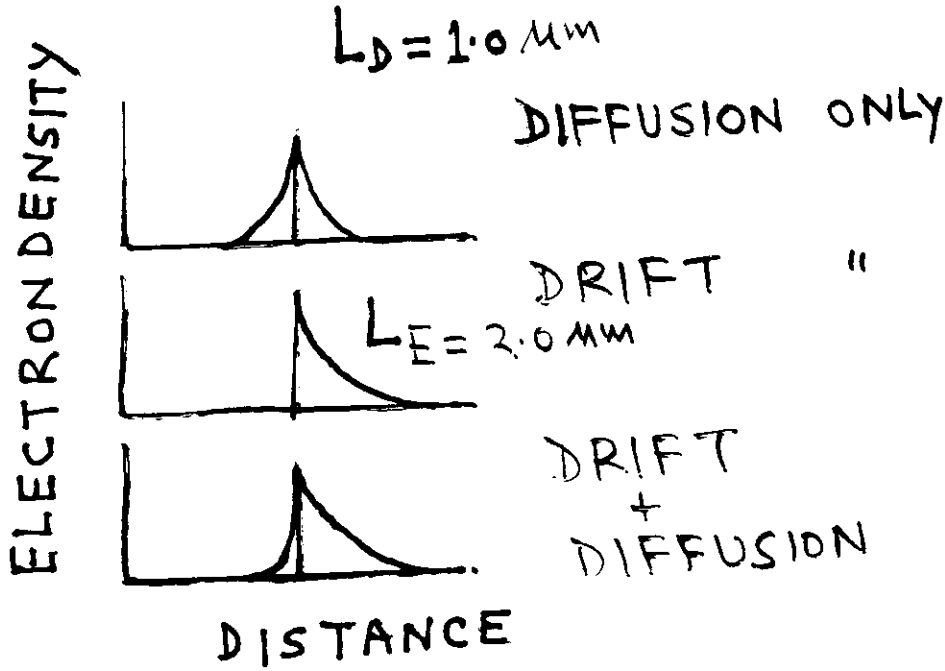
$h\nu + \ominus \longrightarrow \oplus + \bar{e}$

PHOTON DONOR TRAP ELECTRON

RECOMBINATION PROCESS

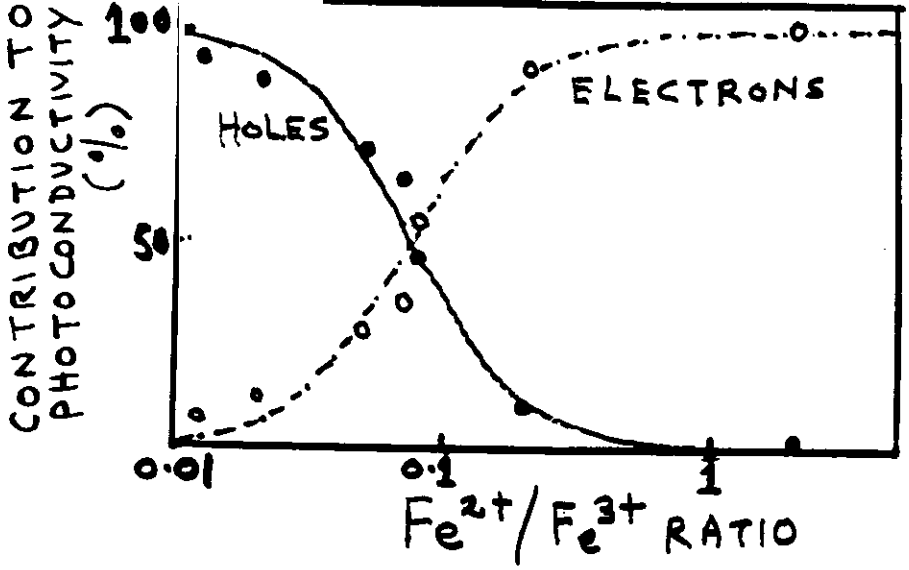
$\oplus + \bar{e} \longrightarrow$ DONOR

TRAP ELECTRON



$L_E = \mu \tau E_0$ → APPL. BIAS ELECTR. FIELD

$L_D = (D\tau)^{1/2} = \left(\frac{k_B T}{e}\right)^{1/2} (\mu\tau)^{1/2}$



$LiNbO_3 : Fe$

—·---- DIRECT PHOTO COND. MEASUREMENTS

O ● HOLOGRAPHIC MEASUREMENTS

- EFFECTIVE TRAP DENSITY — DENSITY OF PHOTOREFRACTIVELY MOBILE CHARGES
- ELECTRO-OPTIC COEFFICIENT — SIGN OF CHARGE CARRIERS.
- CHARGE DIFFUSION LENGTH
- MOBILITIES (TRAPPING)
- RECOMBINATION COEFFICIENTS
- DIELECTRIC CONST.
- BACK GROUND REF. INDEX
- PHOTOIONIZATION CROSS SECTION
- NUMBER DENSITY OF DOPANTS
- NO. DENSITY OF ACCEPTORS THAT COMPENSATE PER NO. DENSITY OF IONIZED DONORS IN DARK.

CHARACTERIZATION

SIMPLE MODEL

- P.R. MEDIA ASSUMED TO CONTAIN CERTAIN TYPE OF IMPURITIES OR IMPERFECTIONS
- ALL DONOR IMPURITIES IDENTICAL, HAVE SAME ENERGY STATE SOMEWHERE IN MIDDLE OF BANDGAP
- DONOR IMPURITIES IONIZED BY ABSORBING PHOTONS GENERATING ELECTRONS IN CONDUCTION BAND LEAVING EMPTY STATES BEHIND. IONIZED IMPURITIES CAPABLE OF CAPTURING ELECTRONS
- RATE OF GENERATION OF ELECTRONS SAME AS THAT OF IONIZED IMPURITIES
- ELECTRONS ARE MOBILE, IMPURITIES STATIONARY
- DENSITY OF DONORS OFTEN MUCH LARGER THAN THAT OF ACCEPTOR IMPURITIES
- ACCEPTOR IMPURITIES FOR PURPOSE OF CHARGE NEUTRALITY ONLY. DO NOT PARTICIPATE (THIS MODEL) IN P.R. EFFECT

KUKHTAREV'S BAND TRANSPORT MODEL: (1979)

RATE EQUATION FOR IONIZED DONOR DENSITY N_D^i

[NEGLECTING THERMAL GENERATION ($\beta \ll sI$)]

$$\frac{\partial N_D^i}{\partial t} = sI\,(N_D - N_D^i) - \gamma_R N N_D^i$$

N_D : DENSITY OF DONOR IMPURITY

N_D^i : DENSITY OF IONIZED DONORS

$(sI+\beta)(N_D-N_D^i)$: RATE OF ELECTRON GENERATION

$\gamma_R N N_D^i$: RATE OF TRAP CAPTURE

N : ELECTRON DENSITY

s : CROSS SECTION FOR PHOTOEXCITATION

I : LIGHT INTENSITY

β: RATE OF THERMAL GENERATION OF ELECTRONS

γ_R: ELECTRON-IONIZED TRAP RECOMBINATION RATE

RATE EQUATION FOR ELECTRON DENSITY N

$$\frac{\partial N}{\partial t} - \frac{\partial N_D^i}{\partial t} = \frac{1}{q}(\nabla \cdot j)$$

j : Current density

$-q$: Electronic charge

CURRENT DENSITY

$$j = qN\mu E + k_B T \mu \nabla N + pI$$

p:equivalent photovoltaic const.

μ : Mobility tensor

k_B:Boltzmann constant

T:Temperature

E: Electric field

POISSON EQUATION

$$\nabla \cdot \epsilon E = \rho(r) = -q\,(N + N_A - N_D^{\,i})$$

$\rho(r)$: Charge density

N_A:Density of acceptor impurity

ϵ : Dielectric tensor

IN THE ABSENCE OF LIGHT ILLUMINATION,

CHARGE NEUTRALITY

$$(N + N_A - N_D^{\,i}) = 0$$

STEADY STATE EQUATIONS:

$$sI\,(N_D - N_D^{\,i}) - \gamma_R N N_D^{\,i} = 0$$

$$\nabla \cdot j = 0$$

$$j = qN\mu E + k_B T \mu \nabla N$$

$$\nabla \cdot \epsilon E = \rho(r) = -q(N + N_A - N_D^{\,i})$$

TRANSIENT SOLUTIONS OF SPACE-CHARGE FIELDS

CONSIDERATIONS OF RATE OF PHOTOEXCITATION, RATE OF RECOMBINATION AND SPEED OF CHARGE TRANSPORTATION ETC.

TIME-DEPENDENT SOLUTIONS
[LOW MODULATION DEPTH ($|I_1| << I_o$)]

$$N(r,t) = N_o(t) + \mathrm{Re}\{N_1(t)\, e^{-iK.r}\}$$

$$N_D^i(r,t) = N_{Do}^i(t) + \mathrm{Re}\{N_{D1}^i(t)\, e^{-iK.r}\}$$

$$j(r,t) = j_o(t) + \mathrm{Re}\{j_1(t)\, e^{-iK.r}\}$$

$$E(r,t) = E_o(t) + \mathrm{Re}\{E_1(t)\, e^{-iK.r}\}$$

WHERE $N_o, N_{Do}^i, j_o, E_o, N_1, N_{D1}^i, j_1, E_1$ ARE CONSTS.

SET OF EQUATIONS FOR ZEROTH AND FIRST ORDER TERMS

$$\frac{dN_{Do}^i}{dt} = sI_o(N_D - N_{Do}^i) - \gamma_R N_o N_{Do}^i$$

$$\frac{dN_o}{dt} = \frac{dN_{Do}^i}{dt}$$

$$j_o = q\mu N_o E_o$$

$$N_{Do}^i + N_o - N_A = 0$$

$$\frac{dN_{D1}^i}{dt} = -(sI_o+\gamma_R N_o)N_{D1}^i - \gamma_R N_{Do}^i N_1 + (N_D - N_{Do}^i)sI_1$$

$$\frac{dN_1}{dt} = \frac{dN_{D1}^i}{dt} - \frac{iK}{q} j_1$$

$$j_1 = q\mu(E_o N_1 + N_o E_1) - ik_B T\mu N_1$$

$$-iK\epsilon E_1 = q(N_{D1}^i - N_1)$$

ϵ IS EFFECTIVE DIELECTRIC CONST. $\langle\epsilon\rangle$
μ IS EFFECTIVE MOBILITY $\langle\mu\rangle$

INTERFERENCE OF TWO WAVES (WAVE-VECTORS $\vec{k}_a$ AND $\vec{k}_b$)

$$I(\vec{r}) = I_o + Re\{I_1 e^{-i\vec{K}.\vec{r}}\}$$

GRATING WAVE-VECTOR $\vec{K}=\vec{k}_b-\vec{k}_a$

FUNDAMENTAL COMPONENT OF SPACE-CHARGE DENSITY

$$\rho = \rho_o \cos(\vec{K}.\vec{r}); \quad \rho_o = \text{CONST.}$$

AMPLITUDE OF SPACE-CHARGE FIELD (POISSON EQN.)

$$E = \rho_o \frac{K}{K.\epsilon K} \sin(\vec{K}.\vec{r})$$

- SHIFT OF $(\pi/2)$ RELATIVE TO INTENSITY PATTERN

SOLUTION OF STEADY STATE EQUATS.

AMPLITUDE OF SPACE-CHARGE FIELD

ASSUMPTIONS:

- LOW MODULATION DEPTH
- $sI_o << \gamma_R N_A$
- $N_D s I_o << \gamma_R N_A^2$

$$E_1 = \frac{iK\frac{k_B T}{q} - \frac{K.\mu E_o}{K<\mu>}}{1 + \frac{K^2}{k_D^2} + i\frac{qK.\mu E_o}{k_B T k_D^2 <\mu>}} * \frac{I_1}{I_o}$$

$$k_D^2 = \frac{q^2}{<\epsilon> k_B T} \frac{N_A}{N_D} (N_D - N_A)$$

$$<\epsilon> = \frac{K.\epsilon K}{K^2} \text{ AND } <\mu> = \frac{K.\mu K}{K^2}$$

PURE DIFFUSION CASE ($E_o=0$)

AMPLITUDE OF SPACE-CHARGE FIELD

$$E_1 = \frac{iK\frac{k_BT}{q}}{1+\frac{K^2}{k_D^2}} * \frac{I_1}{I_o} = \frac{iE_d}{1+\frac{E_d}{E_q}} \frac{I_1}{I_o}$$

$$\text{DIFFUSION FIELD}(E_d) = K\frac{k_BT}{q}$$

$$\text{SATURATION FIELD}(E_q) = K\frac{qN_A}{<\epsilon>K}$$

1. FOR LARGE GRATING SPACING (SMALL K)

$$E_d << E_q \text{ AND } E_1 \approx iE_d \frac{I_1}{I_o}$$

2. FOR SMALL GRATING SPACING (LARGE K)

$$E_q << E_d \text{ AND } E1 \approx iE_q \frac{I_1}{I_o}$$

3. E_1 REACHES A MAXIMUM WHEN $E_d=E_q$

4. FACTOR "i" IN E_1 REPRESENTS PHASE SHIFT OF $(\pi/2)$

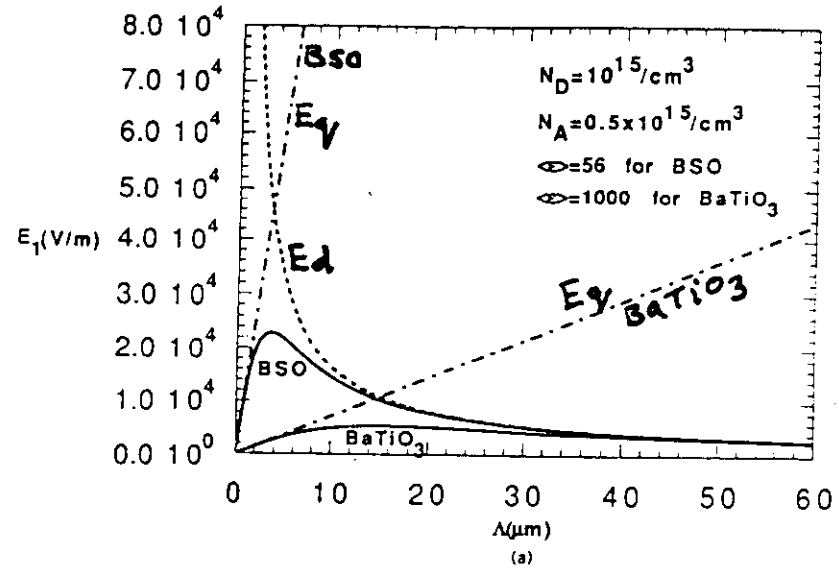
WITH APPLIED FIELD

AMPLITUDE OF SPACE-CHARGE FIELD

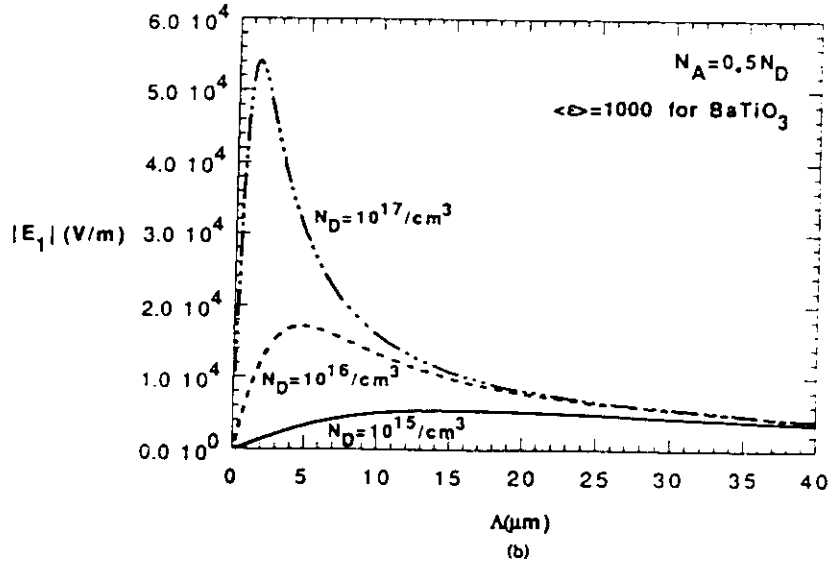
$$E1 = \frac{iE_d}{1+\frac{E_d}{E_q}} \left[\frac{1+\frac{E_o}{E_d}}{1+i\frac{E_o}{(E_d+E_q)}}\right] \frac{I_1}{I_o}$$

SPACE-CHARGE FIELD ($E_o \neq 0$)

TERM IN SQUARE BRACKET
- COMPLEX SCALING FACTOR[AFFECTS MAGNITUDE AND PHASE OF E_1]



SPACE-CHARGE FIELD E_1 Vs GRATING PERIOD Λ FOR $BaTiO_3$ AND BSO



SPACE-CHARGE FIELD E_1 Vs GRATING PERIOD Λ FOR VARIOUS DOPANT DENSITIES

MOVING GRATINGS

INTERFERENCE OF TWO BEAMS (FREQUENCIES ω_a, ω_b)

$$I(\vec{r}) = I_o + Re\{I_1 e^{-i(\Omega t - \vec{K}.\vec{r})}\}$$

$$\Omega = \omega_b - \omega_a$$

AMPLITUDE OF SPACE-CHARGE FIELD

$$E_1 = \frac{iE_d}{1+\frac{E_d}{E_q}} \frac{1+\frac{E_o}{E_d}}{1+i\frac{E_o}{E_d+E_q}} \frac{1}{1+i\Omega t_o \frac{E_d+E_\mu+iE_o}{E_d+E_q+iE_o}} \frac{I_1}{I_o}$$

DRIFT FIELD(E_μ) $= \frac{\gamma_R N_A}{<\mu>K}$ AND $t_o = \frac{N_A}{N_D s I_o}$ AS CHARACTERISTIC TIME CONST. OF MEDIUM

WITHOUT APPLIED FIELD($E_o = 0$)

AMPLITUDE OF SPACE-CHARGE FIELD

$$E_1 = \frac{iE_d}{1+\frac{E_d}{E_q}} \frac{1}{1+i\Omega t_o \frac{E_d+E_\mu}{E_d+E_q}} \frac{I_1}{I_o}$$

COMPLEX AMPLITUDE ALSO WRITTEN AS

$$E_1 = |E_1| e^{i\phi}$$

$$|E_1| = \frac{iE_d}{1+\frac{E_d}{E_q}} \left[\frac{1}{1+\Omega t_o\left(\frac{E_d+E_\mu}{E_d+E_q}\right)^2}\right]^{1/2} \frac{I_1}{I_o}$$

$$\phi = \pi/2 - \tan^{-1}(\Omega t)$$

TRANSIENT SOLUTIONS

SPACE-CHARGE FIELD FORMATION

$$E_1(t) = E_1 \left[1 - e^{-t/\tau}\right]$$

E_1: STEADY STATE SPACE-CHARGE FIELD

AND COMPLEX TIME CONST.

$$\tau = \frac{1 + \dfrac{k_B T \mu K^2}{q\gamma_R N_A} - \dfrac{i\mu K E_o}{\gamma_R N_A}}{1 + \dfrac{K^2}{k_D^2} + i\dfrac{qKE_o}{k_B T k_D^2}} \frac{\epsilon}{q\mu N_o}$$

$$= t_o \frac{E_d + E_\mu + iE_o}{E_d + E_q + iE_o}$$

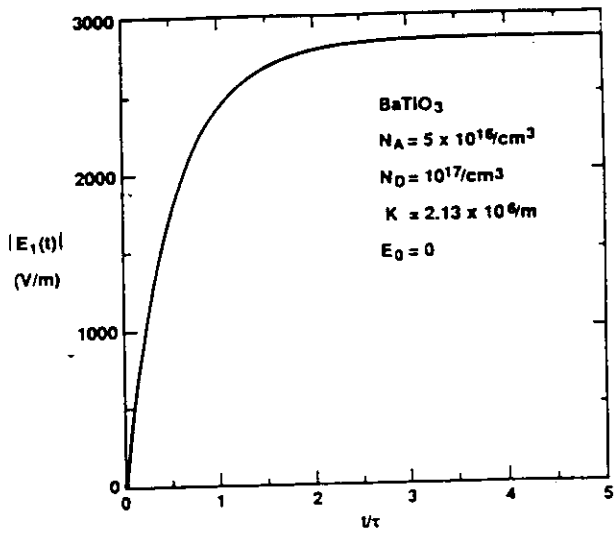
1. WITHOUT APPLIED FIELD [$E_o = 0$], "τ" IS REAL. $E_1(t)$ GROWS EXPONENTIALLY.

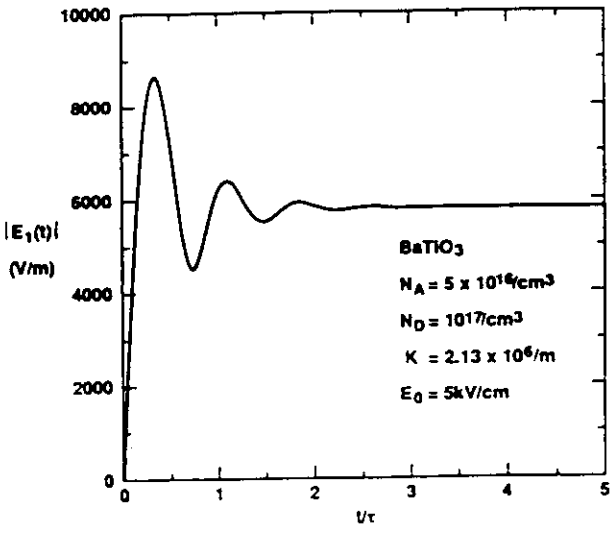
2. WITH FINITE FIELD, "τ" IS COMPLEX. $E_1(t)$ GROWS EXPONENTIALLY WITH OSCILLATIONS.

SPACE-CHARGE FIELD DURING ERASURE

$$E_1(t) = E_1 e^{-t/\tau}$$

- NO OSCILLATIONS DURING ERASURE WITH/WITHOUT APPLIED FIELD


3000
2000
1000
0
$|E_1(t)|$
(V/m)
0
1
2
3
4
5
t/τ
$BaTiO_3$
$N_A = 5 \times 10^{16}/cm^3$
$N_D = 10^{17}/cm^3$
$K = 2.13 \times 10^6/m$
$E_0 = 0$


10000
8000
6000
4000
2000
0
$|E_1(t)|$
(V/m)
0
1
2
3
4
5
t/τ
$BaTiO_3$
$N_A = 5 \times 10^{16}/cm^3$
$N_D = 10^{17}/cm^3$
$K = 2.13 \times 10^6/m$
$E_0 = 5kV/cm$

BAND TRANSPORT MODEL: SHORTCOMINGS

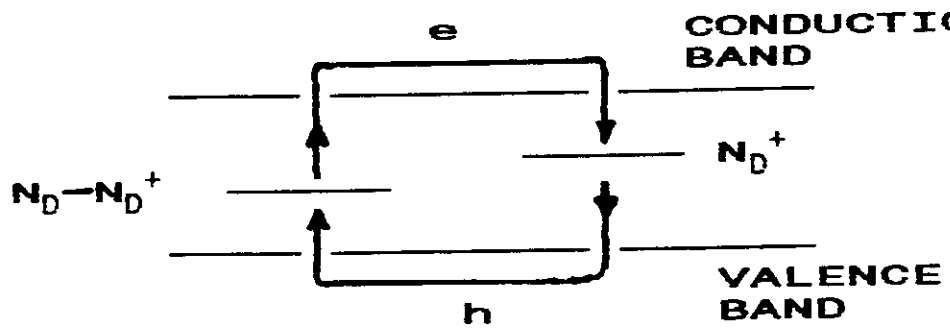
ANOMALOUS EXPERIMENTAL RESULTS [STEADY STATE AND TIME-DEPENDENT BEHAVIOUR OF P.R. MATERIALS]

- SUBLINEAR DEPENDENCE OF PHOTOCONDUCTIVITY/RESPONSE RATE/GRATING ERASURE RATE(τ) WITH INTENSITY ($\tau \propto I^{x}$; $0<x\leq 1$)
- TWO DISTINCT TIME CONSTANTS DURING GRATING DECAY, FOR GRATING FORMATION WITH HIGH INTENSITY PULSED LASERS
- PEAK SPACE-CHARGE FIELD OCCURING AT A LOWER P.R. GRATING SPACING
- NON-EXPONENTIAL DARK DECAYS OF P.R. GRATINGS AND DEPENDENCE OF DARK STORAGE TIME ON THE LIGHT INTENSITY (USED FOR WRITING)
- CHANGE OF SIGN OF COUPLING CONST. (Γ) BY CHANGE OF GRATING WAVE NUMBER/DIFFERENCE IN THE CONCENTRATION OF IONIZED DONORS FOR ELECTRONS AND HOLES
- DECREASE IN DIFFRACTION EFFICIENCY OBSERVED IN BSO
- FIXING AT ELEVATED TEMPERATURE/OR BY PERIODIC POLING FOR LONG-TERM HOLOGRAPHIC STORAGE

EXPLAINED THROUGH INCLUSION OF

- ELECTRON-HOLE TRANSPORT
- MULTIPLE SPECIES OF PHOTO-ACTIVE CENTRES
- MOBILE IONS

I. MODIFIED BAND TRANSPORT MODEL WITH TWO CHARGE CARRIERS AND ONE SET OF RECOMBINATION SITE



$$\frac{\partial N_D^+}{\partial t} = s_e I (N_D - N_D^+) - \gamma_e N_e N_D^+ - s_h I N_D^+ + \gamma_h N_h (N_D - N_D^+)$$

$$\frac{\partial N_e}{\partial t} - \frac{1}{e}(\nabla \cdot j_e) = s_e I (N_D - N_D^+) - \gamma_e N_e N_D^+$$

$$\frac{\partial N_h}{\partial t} + \frac{1}{e}(\nabla \cdot j_h) = s_h I N_D^+ - \gamma_h N_h (N_D - N_D^+)$$

$$j_e = eN_e\mu_e E + k_B T \mu_e \nabla N_e$$

$$j_h = eN_h\mu_h E - k_B T \mu_h \nabla N_h$$

$$\nabla \cdot E = -(4\pi e/\epsilon)(N_A - N_D^+ - N_h + N_e)$$

N_D^+: IONIZED DONOR DENSITY

N_e: ELECTRON DENSITY

N_h: HOLE DENSITY

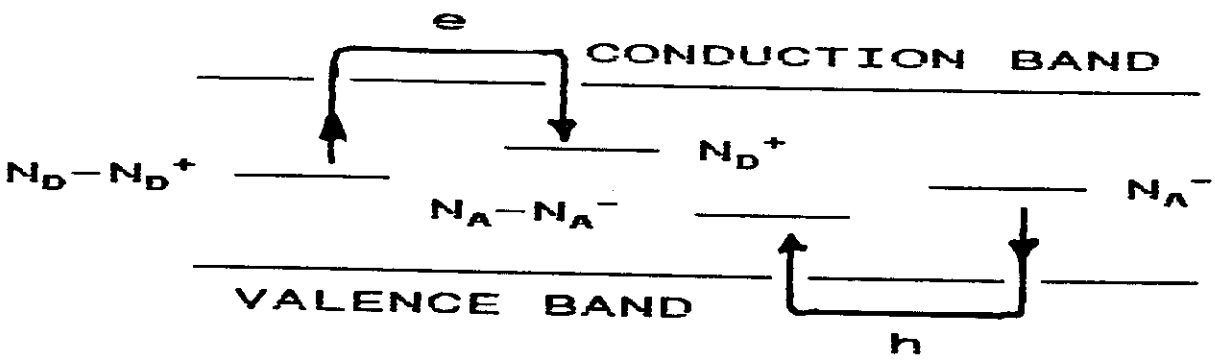
j_e: ELECTRON CURRENT DENSITY

μ_e: ELECTRON MOBILITY

j_h: HOLE CURRENT DENSITY

μ_h: HOLE MOBILITY

II. MODIFIED BAND TRANSPORT MODEL WITH TWO CHARGE CARRIERS AND TWO SETS OF RECOMBINATION SITES



$$\frac{\partial N_D^+}{\partial t} = s_e I (N_D - N_D^+) - \gamma_e N_e N_D^+$$

$$\frac{\partial N_A^-}{\partial t} = s_h I (N_A - N_A^-) - \gamma_h N_h N_A^-$$

$$\frac{\partial N_e}{\partial t} - \frac{\partial N_D^+}{\partial t} = \frac{1}{e} (\nabla \cdot j_e)$$

$$\frac{\partial N_h}{\partial t} - \frac{\partial N_A^-}{\partial t} = -\frac{1}{e} (\nabla \cdot j_h)$$

$$j_e = eN_e\mu_e E + k_B T \mu_e \nabla N_e$$

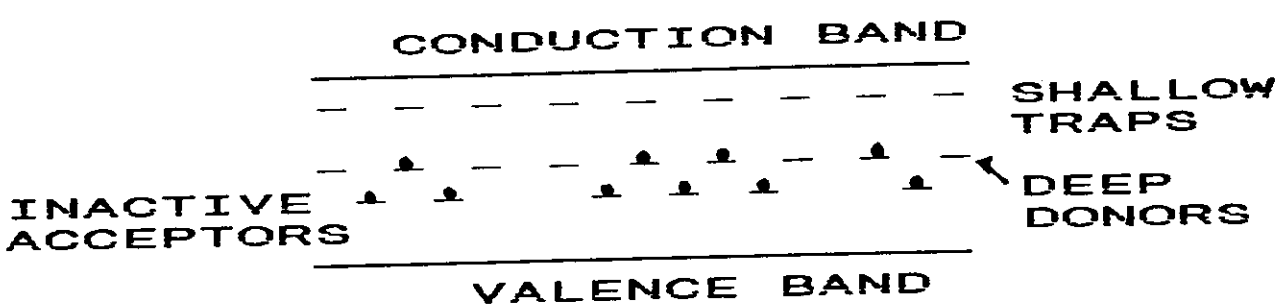
$$j_h = eN_h\mu_h E - k_B T \mu_h \nabla N_h$$

$$\nabla \cdot E = (4\pi e/\epsilon)(N_D^+ - N_A^- + N_h - N_e + \Delta N)$$

$\Delta N = (N_A^- - N_D^+)$ IN DARK

N_A^-: DENSITY OF HOLE DOMINANT LEVELS

III. MODIFIED BAND TRANSPORT MODEL IN PRESENCE OF SHALLOW TRAPS



SHALLOW TRAP MODEL FOR N-TYPE CRYSTALS

$$\frac{\partial N_D^+}{\partial t} = s_D I (N_D - N_D^+) - \gamma_D N N_D^+$$

$$\frac{\partial M}{\partial t} = -(s_T I + \beta) M + \gamma_T N (M_T - M)$$

$$\frac{\partial}{\partial t}(N_D^+ - M - N) + \frac{1}{e}(\nabla \cdot j) = 0$$

$$j = eN\mu E + k_B T \mu \nabla N$$

$$\nabla \cdot E = -(4\pi e/\epsilon)(N - N_D^+ + N_A + M)$$

M: DENSITY OF FILLED SHALLOW TRAPS

M_T: TOTAL SHALLOW TRAP DENSITY

s_D, s_T: LIGHT EXCITATION CROSS-SECT. FOR DONORS AND SHALLOW TRAPS

γ_D, γ_T: RECOMBINATION CONSTANTS FOR DONORS AND SHALLOW TRAPS

β: THERMAL EXCITATION RATE FROM SHALLOW TRAPS

MATERIAL PROPERTIES FOR DYNAMIC HOLOGRAMS.

PHOTOREFRACTIVE SENSITIVITY

DYNAMIC RANGE (MAX. REF. INDEX CHANGE)

PHASE SHIFT BETWEEN REF. INDEX GRATING & INTENSITY GRATING

PHOTOREFR. RECORDING & ERASURE TIME

SPATIAL FREQUENCY DEPENDENCE

ELECTRIC FIELD DEPENDENCE

WAVELENGTH FOR INDUCING REF. INDEX CHANGE

RESOLUTION

SIGNAL-TO-NOISE RATIO

ROOM TEMP. OPERATION

PHOTOREFRACTIVE MATERIALS

THREE SEPARATE CLASSES

- FERROELECTRIC OXIDES
- CUBIC OXIDES OF SILLENITE FAMILY
- SEMI INSULATING COMPOUND SEMICONDUCTORS

FIG. OF MERIT

- MAXIMUM AMPLITUDE OF A REFRACTIVE INDEX HOLOGRAM CAN BE WRITTEN

- IN SILLENITES AND COMPOUND SEMICONDUCTOR ONE MUST APPLY AN EXTERNAL ELECTRIC FIELD TO GET DESIRED R.I. MODULATION

  IN FERROELECTRICS MAXIMUM AMPLITUDE REACHES IN DIFFUSION REGIME

- SILLENITES AND SEMICONDUCTORS HAVE SMALLER INDEX CHANGES

- FERROELECTRICS HAVE LARGER DIELECTRIC CONSTANTS

  ⇒ LARGER DIELECTRIC RELAXATION TIMES ⇒ LARGER RESPONSE TIME

CERAMICS

FERROELECTRIC OXIDES (OXYGEN-OCTAHEDRA)

$LiNbO_3$, $LiTaO_3$, $KTa_{1-x}Nb_xO_3$ (KTN)

$BaTiO_3$, $KNbO_3$, $Sr_{1-x}Ba_xNb_2O_3$ (SBN) → (1.5)

$Ba_2NaNb_5O_{15}$ (BNN) PBN

Typical
$y=0.75$
$x=1.5$
$Ba_{1-x}Sr_xK_{1-y}Na_yNb_5O_{15}$ (BSKNN)

⇒ NO OPTICAL ACTIVITY, HIGHLY BIREFRINGENT

$LiNbO_3$ ⇒ (ILMENITE)

- RESPONSE TIME ~ $10-10^3$ SEC.
- PHOTOREFRACTIVE SENSITIVITY RELATIVELY SMALL, REQD. ENERGY DENSITY ~ 3000 mJ/cm² AT 514.5 nm. (Ar LASER)

$BaTiO_3$ ⇒ (PEROVSKITE)

- RESPONSE TIME ~ 1-10 ms
- MORE SENSITIVE, ENERGY DENSITY ~ 3 mJ/cm² (at 514.5 nm) (Ar LASER)
- LARGE E-O CO-EFFICIENT ($r_{42} \simeq 1640$ pm/V)
- MOST SUITABLE FOR REAL-TIME HOLOGRAPHY
- SELF-PUMPED PC MIRROR

SBN ⇒ (TUNGSTEN BRONZES)

- RESPONSE TIME AND SENSITIVITY SIMILAR TO $BaTiO_3$

ITS LARGEST E-O CO-EFFICIENT IS $r_{33} = 1340$ pm/V

SBN, BSKNN → SELF-PUMPED

$KNbO_3$ ⇒ (SMALL RESPONSE TIME) FASTER
(Perovskite) THAN $BaTiO_3$ AND SBN

- GRATING BUILD UP TIME ~ 100 μS AT 488 nm. of INTENSITY $1 W/cm^2$

* • NO SELF-PUMPED PHASE CONJUGATION OBSERVED

- BEAM COUPLING GAIN CO-EFICIENT GREATER THAN $BaTiO_3$ AND EXCEEDS SELF-PUMPING THRESHOLDS

KTN ⇒ MIXING OF $KNbO_3$ AND $KTaO_3$

- VERY SENSITIVE PR MATERIAL
- ITS AVAILABILITY IS LIMITED BECAUSE OF DIFFICULTIES IN GROWING CRYSTALS OF GOOD OPTICAL QUALITY

COMMERCIAL AVAILABILITY OF MATERIALS WITH LARGE NON-LINEARITIES IS LIMITED.

GROWTH (SIZE & OPTICAL QUALITY) REQUIRES INVESTMENT OF SEVERAL YEARS, SUBSTANTIAL FUNDING AND TALENTED PERSONNEL

CUBIC OXIDES (SILLENITES)

COSTLY

$Bi_{12}SiO_{20}$, $Bi_{12}GeO_{20}$, $Bi_{12}TiO_{20}$

$Bi_4Ge_3O_{12}$: Cr

⇒ NON-BIREFRINGENT IN THE ABSENCE OF ELECTRIC FIELD

HIGLY PHOTO CONDUCTIVE AND OPTICALLY ACTIVE

EXHIBIT ELECTICALLY INDUCED BIREFRINGENCE

$Bi_{12}SiO_{20}$ (BSO) ⇒ E-O COEFFICIENT $r_{41} = 4.5$ pm/V

(6328 Å) OPTICAL ROTATORY POWER 21.4 °/mm

RESPONSE TIME ~ 10^{-3} – 1 SEC

SENSITIVITY ~ 100 MJ/cm² at 514.5 nm

OPERATION AT ELEVATED TEMPERATURE DECREASES RESPONSE TIME AND IMPROVES THE EFFICIENCY

$Bi_{12}TiO_{20}$ (BTO) ⇒ NOT SUITED FOR OPERATION IN THE GREEN, SUITED FOR RED AND POSSIBLY INFRARED

AN AC FIELD USED IN BTO GIVES VERY LARGE GAIN CO-EFFICIENTS

OPTICAL ACTIVITY IS 3.5 TIMES SMALLER THAN BSO/ BGO

SEMI-INSULATING COMPOUND SEMICONDUCTORS

GaAs, CdS, GaAs:Cr, InP:Fe, CdTe

- PEAK SENSITIVITY LIES IN INFRARED
- E-O COEFFICIENTS – SAME MAGNITUDES AS SILLENITES
- ELECTRON AND HOLE MOBILITIES 100 TO 1000 TIMES LARGER THAN OXIDES

CURRENT WORK IN FOUR AREAS

- MOVING GRATING AND AC FIELD TECHNIQUES ⇒ ENHANCE NONLINEARITY
- DOPING TECHNIQUES TO INCREASE DONOR/ACCEPTOR/TRAP DENSITY
- PICO SECOND PULSE OBSERVATIONS
- PHOTOREFRACTIVE EFFECT TO MEASURE PROPERTIES OF MATERIALS

SPATIAL PHASE SHIFT BETWEEN SPACE CHARGE FIELD AND INTERFERENCE PATTERN RANGES FROM 0 TO 90° ⇒ THREE WAYS FOR OPTIMUM ENERGY TRANSFER

. EXTERNAL DC FIELD >> E_q (E_q M. VALUE OF SPACE CHARGE FIELD)

- WEAKER DC AND MOVING IP BY FREQUENCY DETUNING
- AN AC FIELD WITH PERIOD < GRATING FORMATION TIME
- SENSITIVE FOR ENERGY TRANSFER BETWEEN PICOSECOND AND NANOSECOND PULSED BEAMS

$\Delta n = n_i^3 r_{ij} E$

PHOTOREFRACTIVES

OPT. & ELECTRO-OPT. PROPERTIES

MATERIAL	SYMMETRY (POINT GROUP)	E_g OPT. BAND GAP (eV)	r_{ij} at λ (μm)	n_i at λ (μm)	$\in$	$n_i^3 r_{ij}$ [pm/V] FOR MAX. INDEX CHANGE	$\frac{n_i^3 r_{ij}}{\epsilon_i}$ [pm/V] FOR MAX SENSITIVITY	μ (300K) $[cm^2/Vs]$ ELECTRONS	HOLES
III-V SEMICONDUCTORS									
InP	$\bar{4}3m$	1.35	r_{41} = 1.45 (1.06)	3.29 (1.06)	12.6	52	4.1	4600	150
GaAs	$\bar{4}3m$	1.42	r_{41} = 1.2 (1.08)	3.5 (1.02)	13.2	43	3.3	8500	400
GaP	$\bar{4}3m$	2.26	r_{41} = 1.07 (0.56)	3.45 (0.54)	12	44	3.7	110	75
II-VI Semiconductors									
CdTe	$\bar{4}3m$	1.56	r_{41} = 6.8 (3.39)	2.82 (1.3)	9.4	152	16	1050	100
ZnS	$\bar{4}3m$	3.68	r_{41} = 1.2 (0.4)	2.47 (0.45)	16	18	1.1	165	5
ZnSe	$\bar{4}3m$	2.68	r_{41} = 2.0 (0.55)	2.66 (0.55)	9.1	38	4.1		
ZnTe	$\bar{4}3m$	2.27	r_{41} = 4.45 (0.59)	3.1 (0.57)	10.1	133	13		
CdSe	6mm	1.70	r_{33} = 4.3 (3.39)	2.54 (1.15)	10.65	70	6.6	800	
CdS	6mm	2.47	r_{33} = 4 (0.59)	2.48 (0.63)	10.33	61	5.9		

$LiNbO_3$	3m	3.2	$r_{33}=32.2$	(0.63)	2.27 (0.70)	32	320	11
$Bi_{12}SiO_{20}$	23		$r_{41}=5$	(0.63)	2.54	47	82	1.8
$BaTiO_3$	4mm	3.3	$r_{51}=1640$	(0.55)	2.40 (0.63)	3600 / 135	11300 / 387	4.9 / 2.9
			$r_{33}=28$					
$KNbO_3$	4mm	3.2	$r_{33}=64$		2.23	55	690	14
$Ba_{0.25}Sr_{0.75}Nb_2O_6$	4mm	3.2	$r_{33}=1340$	(0.63)	2.30	3400	16300	4.8

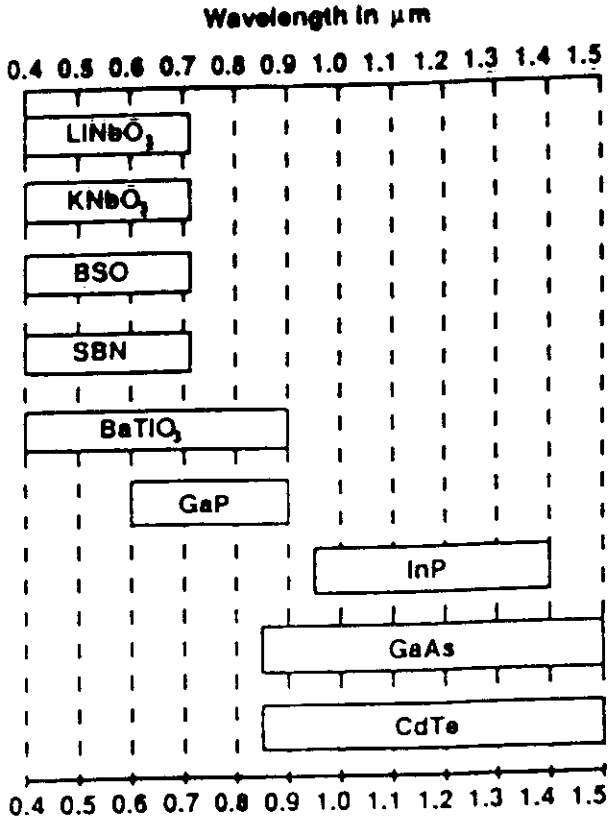
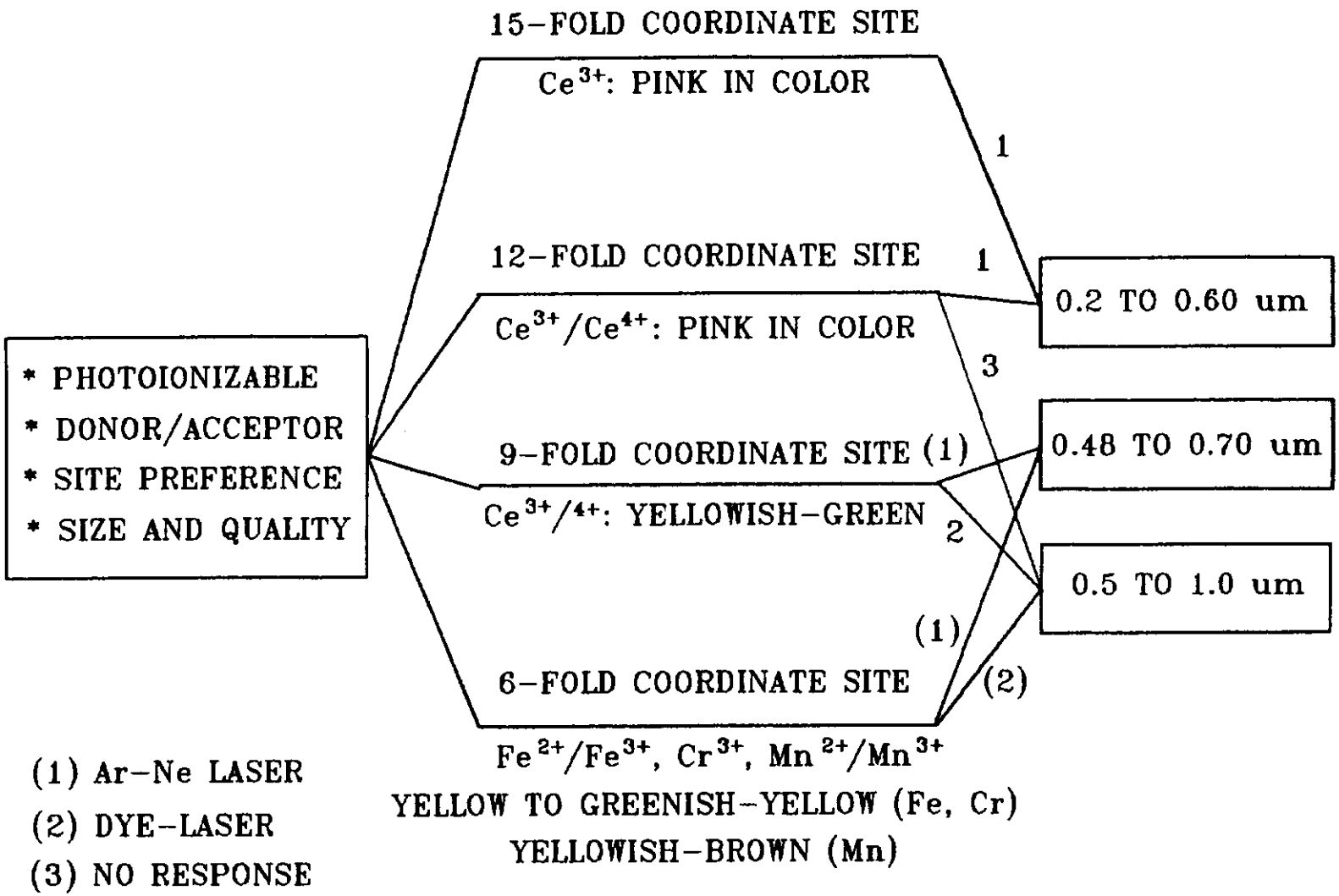


Figure 6. Approximate spectral range of some representative photorefractive materials.

PRELIMINARY PHOTOREFRACTIVE RESULT ON DIFFERENT DOPANTS

PROPERTY	Ce^{3+} -DOPED SBN:60		Cr^{3+}-DOPED SBN:60	Fe^{3+} -DOPED SBN:60
	12-FOLD	9-FOLD	6-FOLD	6-FOLD
CRYSTAL COLOR	PINK	GREENISH-YELLOW	GREENISH-YELLOW	YELLOW
QUALITY	EXCELLENT	EXCELLENT	EXCELLENT	REASONABLE*
ELECTRO-OPTIC COEFFICIENT X 10^{-12} mV	460	460	550	480
BEAM FANNING RESPONSE				
AT 40 mW/cm²	2.5 s	3.0 s	0.7 s	2.8 s
AT 0.2 W/cm²	0.6 s	1.2 s	-	-
AT 2 W/cm²	0.05 s	0.09 s	0.008 s	0.07 s
COUPLING CONSTANT	~19 cm^{-1} (CUBE) ~45 cm^{-1} (PLATE)	~5-6 cm^{-1}	~6-7 cm^{-1}	-
SPECTRAL RESPONSE	0.4-0.7μm	0.4-0.9μm	0.6-1.0μm	0.4-0.9μm
SPPCR	EXCELLENT	EXCELLENT	EXPECTED	EXPECTED

* STRIATED AT HIGHER DOPING LEVELS

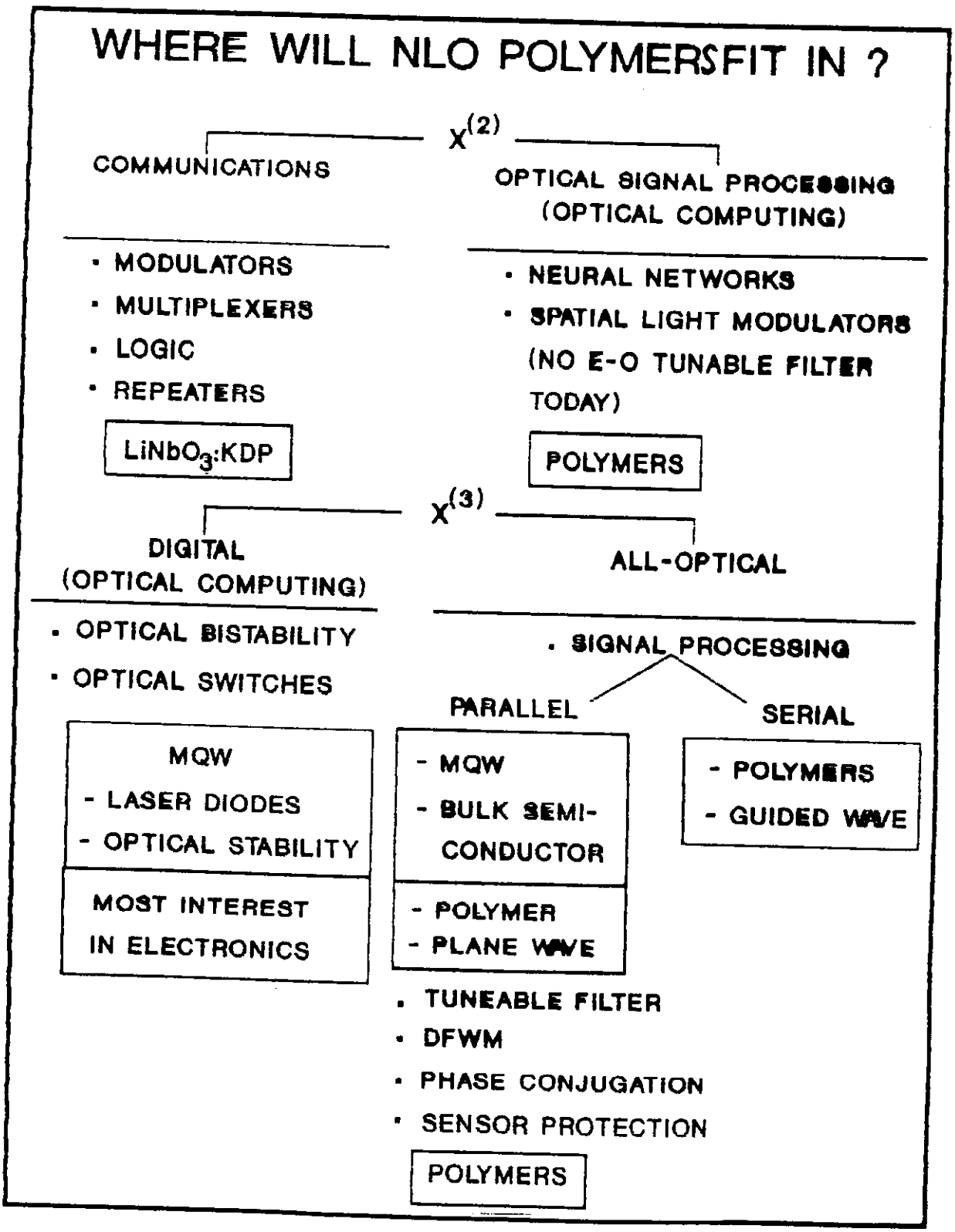


ROLE OF DOPANTS FOR PR APPLICATIONS

WHY NLO POLYMERS ?

- SUBPICOSECOND RESPONSE TIMES
- LARGE, NONRESONANT NONLINEAR ITIES
- LOW DC DIELECTRIC CONSTANTS
- LOW SWITCHING ENERGY
- BROADBAND
- LOW ABSORPTION
- ABSENCE OF DIFFUSION PROBLEMS
- POTENTIAL FOR RESONANT ENHANCEMENT
- EASE OF PROCESSING AND SYNTHESIS MODIFICATION
- ROOM TEMPERATURE OPERATION
- ENVIRONMENTAL STABILITY
- MECHANICAL AND STRUCTURAL INTEGRITY

WHERE WILL NLO POLYMERS FIT IN ?



PHOTOREFRACTIVE POLYMERS:

- BISPHENOL-A-DIGLYCILDYLETHER 4-NITRO-1,2-PHENYLLENEDIAMINE MADE PHOTOCONDUCTIVE BY DOPING WITH HOLE-TRANSPORT AGENT DIETHYL-AMINO-BENZALDEHYDE DIPHENYL-HYDRAZONE [bis A- NPDA:DEH] (DEH IS HOLE-TRANSPORT AGENT TO FACILITATE MONOPOLAR CHARGE TRANSPORT). DIFF.EFF. $\approx 10^{-4}-10^{-5}$.

- METHYL METHACRYLATE COPOLYMER WITH NON-LINEAR CHROMOPHORE p-NITROANILINE IN PENDENT SIDE GROUP DOPED WITH A CHARGE-TRANSPORT AGENT, DIETHYLAMINO-BENZALDEHYDE DIPHENYLHYDRAZONE [PMMA-PNA:DEH].

- FULLERENE MOLECULE C_{60} SENSITIZER FOR PMMA-PNA:DEH.

- METHACRYLIC ESTER POLYMER CONTAINING TRICYANOVINYL-CARBAZOLE GROUP WITH AN ALKYLINE SPACER [5-(N-CARBAZOLYL) PENTYL METHACRYLATE]

- DISPERSE RED (DR1) IN PMMA THIN FILMS [PMMA:DR1].

- 1:1 SIDE-CHAIN COPOLYMER OF N-METHACRYLOXEYETHYL-N-METHYLAMINONITRISTILBENE WITH METHYL METHACRYLATE [DANS/MMA].

- PHOTOCONDUCTING POLYMER POLY(N-VINYLCARBAZOLE), DOPED WITH OPTICALLY NON-LINEAR CHROMOPHORE 3-FLUORO-4-N,N DIETHYLAMINO-β-NITROSTYRENE AND SENSITIZED FOR CHARGE GENERATION WITH 2,4,7-TRINITRO-9-FLUORENONE [PVK:F-DEANST:TNF].

- POLYVINYLCARBAZOLE DOPED WITH NON-LINEAR OPTICAL DYE AND A COPOLYMER OF NLO DYE-PENDANT MONOMER AND MONOMER WITH CARBAZOLYL GROUP 1-CYANO-2-(4-DIMETHYLAMINOPHENYL) ACRYLIC ACID METHYL ESTER AS NLO DYE.

- HOLE TRANSPORTING COMPOUND AND NON-LINEAR OPTICAL CHROMOPHORE COVALENTLY LINKED TO THE POLYMER BACKBONE .

- POLYMER CONTAINING A CONJUGATED BACKBONE AND A SECOND-ORDER NON-LINEAR OPTICAL CHROMOPHORE.

- AZO-DYE-DOPED POLYMER, POLY (VINYLCARBAZOLE): TRINITRO-FLUORENONE/ DISPERSE RED I [PVK-TNF:DRI]. (PHOTOCONDUCTING POLYMER HOST DOPED WITH SMALL CONCENTRATION OF SENSITIZER AND LARGE CONCENTRATION OF NONLINEAR OPTICAL CHROMOPHORE HAVING ORIENTATIONAL MOBILITY AT AMBIENT TEMP).

- NONLINEAR OPTICAL CHROMOPHORE 2,5 DIMETHYL-4-p-NITRO-PHENYLAZOANISOLE DOPED WITH N-ETHYLCARBAZOLE ADDED TO PVK:TNF TO GET HIGH OPTICAL GAIN AND DIFF. EFFICIENCY ≈ 100% [DMNPAA:PVK:ECZ:TNF].

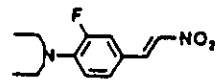
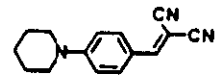
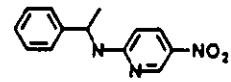
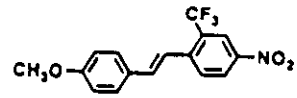
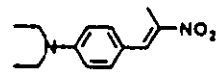
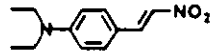
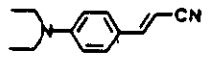
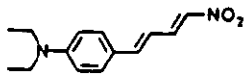
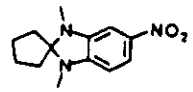
NLO chromophore	Acronym	Structure	α (cm^{-1})	η_{ss}	Γ (cm^{-1})
3-Fluoro-4-*N*,*N*-diethylamino-(*E*)-β-nitrostyrene	FDEANST		1.4	4.5×10^{-4}	7.0
4-Piperidinobenzylidene-malononitrile	PDCST		2.3	3.7×10^{-3}	7.8
(+)-2-(α-Methylbenzyl) amino-5-nitropyridine	MBANP		0.9	1.7×10^{-4}	2.6
4-Methoxy-2'-trifluoro-methyl-4'-nitrostilbene	MTFNS		0.6	1.2×10^{-5}	1.2
4-*N*,*N*-Diethylamino-(*Z*)-β-methyl-(*E*)-β-nitrostyrene	DEANMST		10	1.5×10^{-3}	8.0
4-*N*,*N*-Diethylamino-(*E*)-β-nitrostyrene	DEANST		10	2.3×10^{-3}	5.0
4-*N*,*N*-Diethylamino-(*E*)-cinnamonitrile	DEACST		11	2.0×10^{-4}	2.1
(*E*,*E*)-1-(4-*N*,*N*-Diethyl aminophenyl)-4-nitrobutadiene	DEABNB		40	9.9×10^{-5}	3.2
1,3-Dimethyl-2,2-tetramethylene-5-nitrobenzimidazoline	DTNBI		49	3.8×10^{-4}	5.4

Fig. ●. Nonlinear chromophores that are combined with PVK:TNF at a concentration of 33 wt. %, along with their acronyms, structure, absorption coefficient α, steady-state diffraction efficiency η_{ss}, and two-beam coupling-gain coefficient Γ of the resulting PR materials.