From organic molecules to optical devices

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K. D. Dorkenoo (Optical circuits, data storage, multiphoton microscopy)
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V. Rodriguez (ISM, Bordeaux)
In the field of lasers there are no boundaries, only horizons (Gérard Mourou, 2010).

The business is now at about 7 billion dollars and expanding.

First demonstration of a working laser: Theodore Maiman (1960)

In fact, Gordon Gould probably made the first laser (he finally received his first patent for the laser in 1977).


In the field of lasers there are no boundaries, only horizons (Gérard Mourou, 2010).

Theodore Maiman

Components of the first ruby laser
1961: Real start of Non Linear Optics (NLO)

**Second harmonic generation** (or frequency doubling): process in which a nonlinear optical medium produces light at double the frequency of the input beam.

\[ E^{\omega}, \lambda \quad \rightarrow \quad E^{2\omega}, \frac{\lambda}{2} \]

Quartz crystal

P.A. Franken *et al.* (1961)
Second Harmonic Generation (SHG)


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.
I. Introduction

II. Basic considerations
   - NLO molecular response
   - Push-pull molecules for quadratic optics
   - Quadratic hyperpolarizability optimization
   - Quadratic processes (SHG, TPA) in polymeric materials

III. Examples of applications
   - photopolymerization
   - optical circuits
   - artificial muscles
   - optical data storage

IV. Concluding remarks
Molecular response to a weak E.M. field

Weak field $\rightarrow$ linear response:

Microscopic polarization

$$p_i = \mu_{gi} + \alpha_{ij} e_j$$

Permanent dipole

Induced dipole

$\alpha$ linear polarizability tensor

$e$ local electric field related to external field for a spherical cavity through field factors:

$$f(\omega) = \frac{n?\omega + 2}{3}$$ Lorentz expression ("The theory of electrons," B.G. Teubner ed., Leipzig, 1909)

$$f(0) = \frac{\varepsilon(n^2+2)}{(n^2+2\varepsilon)}$$ Onsager expression (JACS, 58, 1486, 1936)
Molecular response to a strong E.M. field

\[ P_i = \mu g_i + \alpha_{ij} e_j + \beta_{ijk} e_j e_k + \gamma_{ijkl} e_j e_k e_l + \ldots \]

Linear response

Non linear response

\( \beta \): 2nd order polarizability tensor or 1st order hyperpolarizability tensor or quadratic hyperpolarizability tensor

\( \gamma \): 3rd order polarizability tensor or 2nd order hyperpolarizability tensor or cubic hyperpolarizability tensor

\( i, j, k, \ldots \) refer to components expressed in a molecular frame
Macroscopic response

\[
\mathbf{P} = \sum_{\text{molecules}} \mathbf{p} = \sum \mu_{gi}^{i} + \alpha_{ij}^{i} e_{j} + \beta_{ijk}^{i} e_{j} e_{k} + \gamma_{ijkl}^{i} e_{j} e_{k} e_{l} + \ldots
\]

\[
P_{I}(E) = P_{I} + \chi_{I}^{(1)} E_{J} + \chi_{IJK}^{(2)} E_{J} E_{K} + \chi_{IJKL}^{(3)} E_{J} E_{K} E_{L} + \ldots
\]

1, J, K.. components expressed in the laboratory frame

\[
\chi^{(i)} \text{ } \text{i}^{\text{th}} \text{ order susceptibility}
\]

\[
E = E_{0} \cos \omega t, \quad E^{2} = E_{0}^{2} \cos^{2} \omega t = \frac{1}{2}(E_{0}^{2} + E_{0}^{2} \cos 2\omega t)
\]

Dipoles oscillating at 2\(\omega\): Second Harmonic Generation (SHG)

Requirement in the dipolar approximation for quadratic optics:

non-centrosymmetric structures (molecules, functionalized polymers, crystals)

For centrosymmetric systems:

\[
P(-E) = - P(E)
\]

Even order polarizabilities \(\beta, \delta, \ldots\) and even susceptibilities \(\chi^{(2n)}\) \(\{\ldots\} = 0\)
Macroscopic response

For weak fields

\[ \vec{E} \rightarrow \vec{P} = \chi \vec{E} \]

For intense fields

\[ \vec{E} \rightarrow \chi^{(1)} \vec{E} + \chi^{(2)} \vec{E} \vec{E} + \chi^{(3)} \vec{E} \vec{E} \vec{E} + \ldots \]

One-photon absorption

Fluorescence induced by 2-photon absorption

<table>
<thead>
<tr>
<th>Frequency</th>
<th>Frequency of the polarisation</th>
<th>Processes</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \omega_p, \omega_q )</td>
<td>( \omega = \omega_p + \omega_q )</td>
<td>Sum of frequencies</td>
</tr>
<tr>
<td>( \omega_p, \omega_q )</td>
<td>( \omega = \omega_p - \omega_q )</td>
<td>Difference of frequencies</td>
</tr>
<tr>
<td>( \omega_p, \omega_p )</td>
<td>( \omega = 2 \omega_p )</td>
<td>Second Harmonic Generation (SHG)</td>
</tr>
<tr>
<td>( \omega_p, 0 )</td>
<td>( \omega = \omega_p )</td>
<td>Pockels effect</td>
</tr>
<tr>
<td>( \omega_p, \omega_p )</td>
<td>( \omega = 0 )</td>
<td>Optical correction</td>
</tr>
</tbody>
</table>
Molecular compounds for quadratic optics

\[ \vec{P} = \sum_{\text{molecules}} \vec{p} = \sum \mu_{g_i} + \alpha_{ij} e^j + \beta_{ijk} e^j e^k + \gamma_{ijk} e^j e^k e^l + \ldots \]

Need of efficient molecules (with high $\beta$) for quadratic optics:

Dipolar chromophores with asymmetric electronic density distribution of a $\pi$–electron-conjugated bridge obtained with appropriate donor and acceptor moities
Charge transfer molecules: anharmonic oscillators

Asymmetric polarizability of the conjugation path leading to quadratic hyperpolarizability

Induced dipole

Linear response
Non linear response

D
\pi
A
\mu

H₂N – C₆H₄ – NO₂

H₃C – C₆H₄ – N – C₆H₄ – NO₂

H₃C – C₆H₄ – N – C₆H₄ – O

H₂C – C₆H₄ – N – C₆H₄ – NO₂

H₃C – O – C₆H₄ – N – C₆H₄ – O

H₃C – O – C₆H₄ – N – C₆H₄ – CN

H₃C – C₆H₄ – N – C₆H₄ – CN

H₃C – C₆H₄ – N – C₆H₄ – CN

H₃C – C₆H₄ – N – C₆H₄ – CN
Molecular origin of the optical properties of organics

Orientation

Anisotropy of the linear polarizability
\( \mu^2 \Delta \alpha \)
Refractive index modulation through \( \chi^{(1)} \)
Refractive polymers

Hyperpolarizability

Quadratic hyperpolarizability
\( \mu \beta \)
Refractive index modulation through \( \chi^{(2)} \)
E.O. materials

Donnor
Accepter

\( E \)
Determination of the quadratic hyperpolarizability

GOAL: Guides for the synthesis of very efficient molecules with giant quadratic hyperpolarizabilities

Methods

Finite-Field (FF) method: calculates the ground state dipole in the presence of a static electric field (M. J. S. Dewar and J. P. P. Stewart, Chem. Phys. Lett., 11, 416, 1984)

\[ \alpha_{ij} = \left. \frac{\partial \mu^i_g}{\partial e_j} \right|_{e=0} \]

\[ \beta_{ijk} = \beta_{ikj} = \left. \frac{1}{2!} \frac{\partial^2 \mu^i_g}{\partial e_i \partial e_k} \right|_{e=0} \]


\[ \beta \propto \frac{\mu_{01}^2 \Delta \mu_{01}^2}{(E_{01} - \hbar \omega - i\Gamma_{01})^2(E_{01} - 2\hbar \omega - i\Gamma_{01})} + \frac{\mu_{01}^2 \Delta \mu_{01}^2}{(E_{01} - \hbar \omega - i\Gamma_{01})^2(E_{01} - 2\hbar \omega - i\Gamma_{01})} - \frac{\mu_{01}^2 \Delta \mu_{01}^2}{(E_{01} - \hbar \omega - i\Gamma_{01})^2(E_{01} + \hbar \omega + i\Gamma_{01})} \]

\( \mu_{01} \) transition dipole between states \( S_0 \) and \( S_1 \),
\( \Delta \mu_{01} \) difference between the permanent dipolar moment of \( S_0 \) and \( S_1 \),
\( \Gamma_{01} \) damping factor of \( S_0 \)
Evaluation of the quadratic hyperpolarizability of push-pull molecules

Semi-empirical approaches:

- A. Two-level approximation (rod-like molecules)

\[ \beta_{zzz}(-2\omega; \omega, \omega) = \frac{\omega_{eg}^4}{(\omega_{eg}^2 - 4\omega^2)(\omega_{eg}^2 - \omega^2)} \frac{3}{2} \frac{\mu_{eg}^2 \Delta \mu}{(\hbar \omega_{eg})^2} \]

\[ \beta_{zzz}(-2\omega; \omega, \omega) = F(\omega) \beta(0) \]

\[ \beta(0) = \frac{3}{2} \frac{\mu_{eg}^2 \Delta \mu}{(\hbar \omega_{eg})^2} \]

\[ \Delta \mu = \mu_e - \mu_g \]
-B. Bond Length Alternation (polyenes)
(S. Marder, et al., JACS, (1994) 116, 23,10708)

BLA = average length difference between adjacent carbon-carbon bonds

Neutral form

Zwitterionic form

-C. Two-level two-form approximation (push-pull molecules)


2 forms

\[
\begin{align*}
\text{D} & \rightarrow \text{A}^+ \\
\text{N} & \rightarrow \text{Z}^-
\end{align*}
\]

2 levels

\[
\begin{align*}
|e\rangle &= -\sin\frac{\theta}{2}|N\rangle + \cos\frac{\theta}{2}|Z\rangle \\
|g\rangle &= \cos\frac{\theta}{2}|N\rangle + \sin\frac{\theta}{2}|Z\rangle
\end{align*}
\]

\[
\tan \theta = \frac{2t}{V}
\]

\[
\text{MIX} = -\cos \theta = -\frac{V}{\sqrt{V^2 + 4t^2}}
\]
Experimental determination of the quadratic hyperpolarizability

**EFISH** (Electric Field Induced Second Harmonic) technique:


Determination of both sign and magnitude of scalar product $\mu_g\beta(2\omega)$ with respect to a quartz crystal by **SHG** detection

- Polar molecules in solution
- Strong static electric field applied to break the centrosymmetry
- Need to be away from resonances at $\omega$ and $2\omega$ (usual operating wavelength at 1.907 nm)

**EOAM** (Electro Optical Absorption Measurements)


**Measurement of the electric-field-induced shift of the absorption spectra**

- Polar molecules
- Well suited for elongated chromophores
- Determination of $\Delta\mu$ more reliable than solvatochromism

**HRS** (Hyper Rayleigh Scattering) technique


**Relative measurement of the magnitude of $\beta$**

- Available for ionic and non polar molecules
- Incoherent light-scattering detection
- Operating fundamental wavelength usually at 1064 nm
- Problems correlated to two-photon absorption induced fluorescence
From microscopic to macroscopic: Need of a molecular alignment

Polymer functionalized with NLO chromophores

Without poling

\[ I^\omega, \lambda \rightarrow \text{NO SHG} \]

\[ I^\omega, \lambda \rightarrow \text{SHG with molecular alignment} \]

\[ E_0 \]

Polymers functionalized with NLO chromophores.
Electro-optic effect in organics

\[ P_1(E) = P_i + \chi^{(1)}_{ij} E_j + \chi^{(2)}_{ijk} E_j E_k + \chi^{(3)}_{ijkl} E_j E_k E_l + \cdots \]

\( \chi^2(\omega) \): SHG, Sum- and difference- frequency, Pockels effect, related to molecular parameters by:

\[ \chi^2(\omega) = N \beta(\omega, \varepsilon) < \cos^3 \theta > f(\omega) \]

\( \theta \) average molecular orientation relative to the electric field

Dominant linear Pockels EO effect tensor \( r_{33}(\omega) = \frac{-2 \chi^{(2)}_{zzz}(\omega)}{n^4} \) (in pm/V)

Gas phase approximation (non-interacting dipolar chromophores)

\[ \langle \cos^3 \theta \rangle = L_3(g) \]

\( g = \mu E / kT \)

and for \( g << 1 \),

\[ \langle \cos^3 \theta \rangle = \mu E / 5kT \]
Strategies for the optimization of the NLO properties of polymers with embedded push-pull molecules

1. Increase of $\beta$ and/or $\mu\beta$
   - Lot of papers to optimize the charge transfer through optimized electron donors and electron acceptors substituents and aromatic bridges.
   - Giant hyperpolarizability obtained using twisted intramolecular charge transfer chromophores with $\mu\beta$ around $500\ 000\ 10^{-48}$ esu (nearly $10^3\times$DR1, a model compound!)

2. Increase of N
   - Shape engineering to reduce dipole-dipole interactions
   - Use of dendrimetric structures or hyperbranched polymers
   - Association of chromophores incorporated using specific chemical tools (Click chemistry, RAFT, ATRP) with adapted matrices

3. Optimization of $\cos^3\theta$
   - Increase of the poling field strength by reducing chromophore conductivity
   - Laser-assisted electric field poling (using cis-trans photoisomerisation mechanisms)
   - Corona poling in adapted poling cell
Corona poling cell

- Laser source
- Corona grid
- SHG
- Photomultiplier
- Gas input
- Corona tension max = 10 kV
- Corona current > 100 μA
- Temperature $T_{amb.} \rightarrow 200°C$
- Vacuum ≈ 20 mBars
- PMMA doped with DR1 molecules

$\frac{I(2\omega)}{I(2\omega)_{max}}$

from L. Mager, IPCMS
Quadratic NLO efficiencies


Two-photon absorption (TPA)

(a) Single photon absorption
(b) Two-photon absorption (TPA)
(c) Second harmonic generation (SHG)

Experimental demonstration:

Theoretical prediction of TPA:
M. Göppert-Mayer, Naturwissenschaften 1929, 17, 932 (1929),
Ann. Phys. 9, 273 (1931)
TPA at the microscopic scale

SOS perturbative approach, 3 level contributions

\[
\sigma_{2\text{photons}} = \frac{3}{10} \frac{\hbar \omega^2}{\epsilon_0 c^2 n^2} f^4 \text{Im} < \gamma(-\omega; \omega, \omega, -\omega) >
\]

\[
\gamma \propto \frac{\mu_{01}^2 \Delta \mu_{01}^2}{(E_{01} - \hbar \omega - i \Gamma_{01})^2 (E_{01} - 2\hbar \omega - i \Gamma_{01})} + \frac{\mu_{01}^2 \mu_{12}^2}{(E_{01} - \hbar \omega - i \Gamma_{01})^2 (E_{02} - 2\hbar \omega - i \Gamma_{02})} - \frac{\mu_{01}^4}{(E_{01} - \hbar \omega - i \Gamma_{01})^2 (E_{01} + \hbar \omega + i \Gamma_{01})}
\]

\[
\text{proba}_{2\text{photons}} \approx \frac{\sigma_{2\text{photons}}}{f_r} \times \frac{<P>^2}{\tau S^2}
\]

\[
\begin{align*}
\sigma_{2\text{photons}} & : \text{TPA cross section} \\
f_r & : \text{laser frequency} \\
P & : \text{laser power} \\
S & : \text{surface} \\
\tau & : \text{impulsion duration}
\end{align*}
\]

Need of a high spatial density of photons during a short time

→ femtosecond laser beams
Macroscopic approach

\[ \bar{P}_\omega^{(3)} = \sum_i \bar{e}_i \left( \sum_j \sum_k \sum_l \chi_{ijkl}^{(3)}(\omega, \omega, -\omega) E^j_\omega (E^k_\omega)^* E^l_\omega \right) \]

Here the third-order susceptibility \( \chi^{(3)} \) is given by:

\[ \chi_{ijkl}^{(3)} = N f_2 < \gamma_{ijkl} > \]

\( \chi^{(3)} \) related to two-photon absorption and to the optical Kerr effect

For theoretical considerations, see for example:

The propagating solution can be expressed as follows:

\[ E(z,t) = A(z) \exp \left[ i(kz + \phi(z) - \omega t) \right] = A(z) \exp \left[ i \left( \frac{\omega n_0}{c} + \frac{3\omega}{8cn_0} \chi_r^{(3)} |A|^2 z - \omega t \right) \right] \]  

(31)

This last equation shows that the wave is propagating with a wave number:

\[ k = \frac{\omega}{c} \left( n_0 + \frac{3}{8n_0} \chi_r^{(3)} |A|^2 \right) \]  

(32)

which can be seen as equivalent to having a refractive index \( n \):

\[ n = n_0 + \frac{3}{8n_0} \chi_r^{(3)} |A|^2 \]

\[ = n_0 + \frac{3}{4n_0^2 \varepsilon_0 c} \chi_r^{(3)} I \]  

(33)

By introducing:

\[ n_2 = \frac{3}{4n_0^2 \varepsilon_0 c} \chi_r^{(3)} \]  

(34)

we can write:

\[ n(I) = n_0 + n_2 I \]  

(35)


Gaussian beam in a nonlinear media:
(a) self-focusing, (b) self-defocusing
Voxel size with TPA

Influence of the numerical aperture (NA=nsinθ)

\[ \omega_{xy} = \begin{cases} \frac{0.32\lambda}{\sqrt{2N.A.}} & \text{if N.A.} \leq 0.7 \\ \frac{0.325\lambda}{\sqrt{2N.A.}^{0.91}} & \text{if N.A.} > 0.7 \end{cases} \]

and \[ \omega_z = \frac{0.532\lambda}{\sqrt{2}} \left( \frac{1}{n - \sqrt{n^2 - N.A.}^2} \right) \]

\[ V_{2\text{photons}} = \pi^{3/2} \omega_{xy}^2 \omega_z \]


### Table

<table>
<thead>
<tr>
<th>N.A</th>
<th>( \omega_{xy} ) (μm)</th>
<th>( \omega_z ) (μm)</th>
<th>( V_{2\text{photons}} ) (μm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.3</td>
<td>0.92 (0.68)</td>
<td>9.65 (11.23)</td>
<td>45.5</td>
</tr>
<tr>
<td>0.6</td>
<td>0.46 (0.34)</td>
<td>2.42 (2.71)</td>
<td>2.9</td>
</tr>
<tr>
<td>0.85</td>
<td>0.32 (0.24)</td>
<td>1.23 (1.29)</td>
<td>0.7</td>
</tr>
<tr>
<td>1.3</td>
<td>0.21 (0.16)</td>
<td>0.51 (0.45)</td>
<td>0.13</td>
</tr>
</tbody>
</table>
Two-photon induced fluorescence

Excited volume $< 1 \mu m^3$

$I_{\text{fluorescence, unphotons}} \propto I_{\text{excitation}}$

$I_{\text{fluorescence, 2photons}} \propto I_{\text{excitation}}^2$

Ar laser (514,5 nm)

Ti:sapphire laser (800 nm)

photo: Brad AMOS MRC, Cambridge
Applications of quadratic processes in organics

3D Micro-fabrication and lithography
Photopolymerization
Optical circuits
Organic lasers
Photodynamic therapy
Imaging
High frequency modulators
High density optical storage

... (like a “Prévert list”)
Micro- and nano-fabrication through two photon absorption polymerization: Pioneering works


P. N. Prasad et al., Applied Physics Letters, 74, 170, 1999
3D micro-structures


Nowadays, spatial resolution around 60 nm!
Photopolymerized waveguides

Self-focusing and self-trapping of optical beams upon photopolymerization

Integration of micrometer-sized polymer elements at the end of optical fibers by free-radical photopolymerization
Light induced self-written (LISW) waveguides

A. Monomode guide

$\lambda = 514\,\text{nm}$

**Monomode optical fiber**
Cladding: 125$\mu$m, Core 3$\mu$m

$P = 5\,\mu W$

Single waveguide

$P = 16\,\mu W$

Chaotic behaviour
Solitonic propagation

Linear process: 
Diffraction (dispersion)

Non-linear process: self-focusing due to photopolymerization (self-phase modulation)

linear = non-linear → solitonic behaviour

\[ ik_0 n_0 \frac{\partial E}{\partial z} + \frac{1}{2} \left( \frac{\partial^2 E}{\partial x^2} + \frac{\partial^2 E}{\partial y^2} \right) + k_0^2 n_0 \Delta n E + \frac{i}{2} k_0 n_0 \alpha E = 0 \]
Light induced self written waveguides (LISW)

\[ V = \frac{2\pi}{\lambda} a \sqrt{n_{\text{core}}^2 - n_{\text{cladding}}^2} \]

2a: diameter of the core

Solitonic propagation if V < 2.4

Simulations

Experiments

\[ V = 20 \]

\[ V = 5 \]

\[ V = 1.5 \]

Examples of applications

Fibres connections

Long (\# 10 cm) and flexible guides

Integrated components

Multimode waveguides

\[ \lambda = 514 \text{ nm} \]

**Optical multimode fiber**
Cladding: 250 μm, Core 62.5 μm

Filamentation at a multimode optical fiber exit

Self-written waveguide created using a white light from a 500W halogen lamp
Building of a LISW guided “on the flow”
Multimode fiber connections

Functionalized LISW

LISW functionalized with photochromic molecule


LISW functionalized with dye for laser operation


LISW functionalized with 5CB molecule for phase modulation

TPA set-up for 3D controlled polymerization
TPA polymerization: fibres connection

Cell

Fiber
- \( \phi \text{ core} = 3 \, \mu m \)
- \( \phi \text{ cladding} = 125 \, \mu m \)

Guide created by TPA polymerization

HeNe laser beam

0.8 mm
Fibres connection through a curved guide

1.3 mm

1.5 mm

0.15 mm
Passive Mach-Zehnder

Active E.O. modulator

EO polymer optical waveguide
Traveling-wave electrodes
Substrate

• Artificial Muscles
Artificial Muscles

Liquid crystal elastomer

monomer acrylate

\[
\begin{align*}
\text{C}_4\text{H}_9\text{O} &- \text{C} \rightarrow \text{O} & \text{C} & \text{O} \\
\text{(CH}_2\text{)}_4 & \text{O} & \text{OC}_4\text{H}_9
\end{align*}
\]

Crosslinker 1,6-hexanediol diacrylate

UV photoinitiator 2-benzyl-2-(dimethylamine)-4'-morpholinobutyrophenone (Irgacure 369)

Photopolymerisation and temperature induced reversible contraction:

Aligned sample
Thermoactive microsystems for diffractive optics

30 µm period grating on 30 µm thick sample
Optics Express, 15, 6784 (2007)
• Optical Data Storage
Commercially available optical disks

CD: $\lambda = 780$ nm
track spacing 1.6 $\mu$m.

700 Mo

DVD: $\lambda = 650$ nm
track spacing 740 nm.

5 Go

Blu-Ray: $\lambda = 405$ nm
track spacing 300 nm

25 Go (1 layer) or
50 Go (2 layers)

Need to increase the storage capacity
Roadmap for optical storage technologies
© Technical University Berlin
Organic optical memories: previous studies

3D optical storage, 2 photon writing, reading and erasing of the information (fluorescence) using photochromic molecules embedded in a polymer matrix

3D optical data storage, 2 photon excitation ($10^{12}$ bits/cm$^3$) for photopolymerization

All optical poling, orientational hole burning and trans to cis isomerization of azo-dye chromophores, SHG detection

Two photon memory recording of 100 planes of 30µmx30µmx80µm

D-$\pi$-D molecules for 2 photon polymerization for 3D optical data storage

3D optical data storage in microfibers of polymers, superposition of phase gratings in the microfibers
Organic optical memories: previous studies

Gang Xu et al., Optics communication, 88 (1999)
All optical poling for orientational hole burning and photoinduced reorientation of azobenzene molecules.

P. N. Prasad et al., APL 74, 1338 (1999)
3D optical data storage using 2 photon up converting chromophore, series of images separated by 6µm in depth.

TPA + Nonlinear hole burning in PMMA-DR1.

J. Zyss et al. Optics Express, 13, 505, 2005
 Encoding of nonlinear information by all-optical poling of photoisomerizable NLO molecules, 2µm resolution.

W. Yuan et al., Advanced Materials, 17, 156 (2005)
TPA and photochromic materials (spiropyran) for 3D memory systems, 4µm resolution.

TPA energy transfer enhanced using quantum dot and azo-dye doped polymers.

Five –dimensional optical recording mediated by surface plasmons in gold nanorods (Tera bits storage).
High density data storage

PMMA DR1 sample

Photo-isomerization cis-trans

Step 1: Obtention of an aligned sample by corona poling

Step 2: Local disorientation of DR1 molecules by two photon absorption (photoisomerisation cycles)

Step 3: Reading of the data by SHG

3D SHG grating. Smallest distance between two adjacent points (column on the left hand side) is about 2 μm (writing power 23 mW, exposure time 200 ms with objectif of NA: 0.85, reading power 1.8 mW, scanning speed 10 μm/s). $10^{12}$ dots/cm$^2$. 4.5 µm Data inscription
Rewritable Data Storage

(a) SHG image taken at 3 mW, 785 nm, 10 μm/s, of a grating (50 lines of 50 μm with a pitch of 4μm) written at P = 10 mW, 800 nm (a border burned in at 105 mW serves as a marker); (b) microscope image of the same area; (c) SHG scan after repoling; (d) SHG scan after rewriting 20 lines of 50 μm with a pitch of 10 μm.

Optical and AFM images of printed dots. (a) Optical microscopy of an array of 20 lines of dots written with illumination power increasing from 1.3 (top left) to 48.1 mW (bottom right). (b) SHG imaging scan of the same area of the sample with a power of 2 mW. (c)–(e) AFM scans of three areas depicted by white squares in hole depths of (c) 4, (d) 7, and (e) 20 nm. Scale bars in all pictures are 7.5 μm.

Optics express, 14, 9896 (2006)
JOSA B, 24, 532 (2007)
Gray Scale Imaging

- **a-** Initial image of M. Göppert-Mayer
- **b-** Optical microscopy image
- **c-** SHG image

(a) Initial image of M. Göppert-Mayer, (b) Optical microscopy image, (c) SHG image

Adv. In Opto Electronics, D967613,(2009)
As a conclusion……..

“Physics would be dull and life most unfulfilling if all physical phenomena around us were linear. Fortunately, we are living in a nonlinear world. While linearization beautifies physics, nonlinearity provides excitement in physics.”

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