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The Nonlinear Optics Group (NLOG) at Soreq NRC is engaged in the development of fundamental and applied technology in the related fields of nonlinear optics and laser development. Our work in nonlinear optics started with the goal of improving laser performance. These efforts were successful and opened the way for R&D in nonlinear optics for other applications. Today we use nonlinear optics to enable continuous tunability of lasers, control the path of light beams, modulate a light signal rapidly, provide optical data storage, and supply new means of microscopically probing biological and inorganic samples. Technology maturation and interaction with users will show which aspects of nonlinear optics will make the most impact.

Activities to be discussed in this report include:

1. Development of flashlamp, diode-laser and laser pumped solid-state lasers.
2. Optical parametric oscillators and sum frequency mixing devices for coherent light generation throughout the visible and infrared portions of the spectrum. These devices are based on the application of commercial single domain crystals and on Soreq fabricated periodically poled bulk and waveguide confined crystals.
3. Nonlinear optical materials development with the goal of designing organic materials which exhibit nonlinear absorption (reverse saturable absorbers), electro-optic polymers, and lasing materials for use in disordered highly scattering media.
4. Development of remote sensing systems based on the technologies developed by the group.
5. Subwavelength nonlinear microscopy effects.

Project sponsors include the Israeli and United States Governments and Israeli private industry. Inter alia, commercial products have been developed.

# ***Advances in Nonlinear Optics and Laser R & D***

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## ***1. Summary***

## **2. Laser development**

### **2.1 Flashlamp pumped lasers**

One of the group's long term goals is the development of high energy compact laser systems. To put this into perspective, the aim is to build shoe-box-size lasers that emit up to 10 Joules, and table-top lasers that produce up to 1000 J. In all cases, generation of a high quality laser beam is a must. High laser output implies even higher pump energies. Flashlamp pumping of high energy solid-state lasers is widely used because of its low cost and easy scalability to high energy.

We are well on the way to reaching the goal of developing high energy solid-state lasers. To a large extent, our efforts were successful due to the application of unique designs and to the incorporation of nonlinear and adaptive optics devices [Phase Conjugate Mirrors (PCMs) and Variable Radius Mirrors (VRMs)]. These devices allowed use of laser materials with improved storage capacity and high efficiency, such as Nd:Cr:GSGG (Gadolinium Scandium Gallium Garnet) and Nd:glass which, due to poor thermo-optical characteristics, were previously by relegated to curiosity driven research devices or to low average power systems.

To date, 1.7 J pulse energy at 7 W average power and 4 J at 1 W have been obtained in GSGG and glass lasers.<sup>(1,2)</sup> Higher energies are expected, the limiting factor being damage in polarization rotating Faraday rotators. A test program showed an anomalous pulse duration scaling for Terbium (Tb) doped materials and that switching from Terbium Gallium Garnet (TGG) to Tb:glass increased the damage threshold fivefold (Fig.1).<sup>(2)</sup> This data is a highly significant for high fluence laser designs.

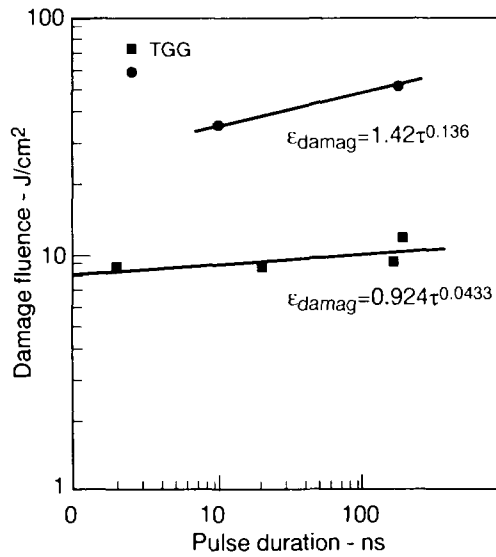


Fig. 1: Faraday rotator damage threshold dependence on pulse duration and material.

The high energy laser design uses a low power oscillator followed by a high power multiple-pass amplifier (MPA). At present, the MPAs are four-pass devices incorporating a VRM and PCM to correct for lowest and higher order thermal lensing aberrations, and Faraday rotators to correct for birefringence and to provide beam path control and isolation (Fig. 2).

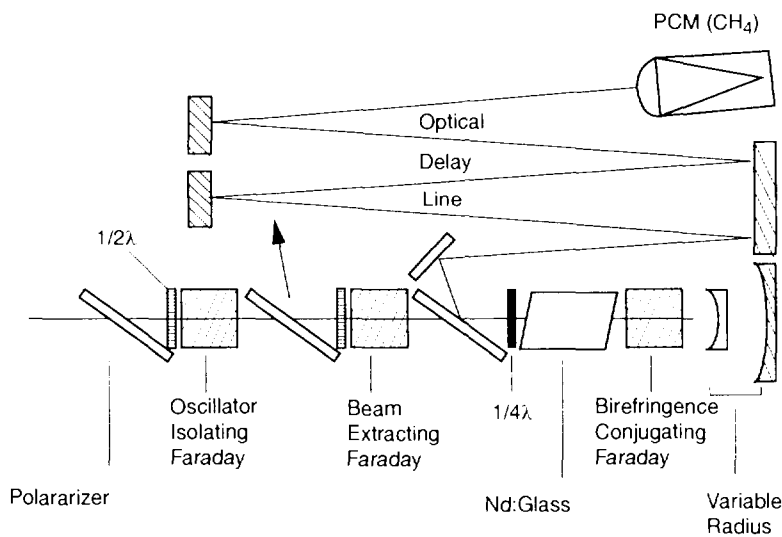


Fig. 2: Schematic design of a high energy quadruple-pass phase and polarization conjugated multiple-pass amplifier.

## 2.2 Flashlamp pumped oscillator development

Development of Nd:Cr:GSGG and Er:Yb:Cr glass oscillators is motivated by the twofold better efficiency of Nd:Cr:GSGG as compared with Nd:YAG, and by the 1535 nm “eye-safe” wavelength of Er:glass. The thermal aberrations and birefringence encountered in MPAs are even more critical in oscillators, due to the larger number of passes through the distorting medium. We concentrated on the application of VRMs to correct for thermal focusing, since PCMs have not proven effective in oscillator applications. The VRM is an adaptive optic with one degree of freedom, and can be used effectively to compensate for changes in rod thermal focusing for radii of curvature of  $\infty$  to 50 cm simply by changing the spacing between two optical elements. Figure 3 compares the performance of a hemispherical GSGG oscillator with a conventional mirror and with a VRM. The much broader operating range of the laser with the VRM is quite pronounced. Complete microprocessor-controlled feedback makes the VRM an adaptive optic.

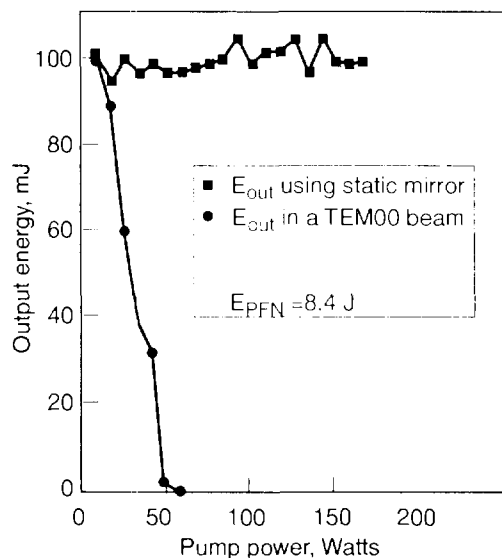
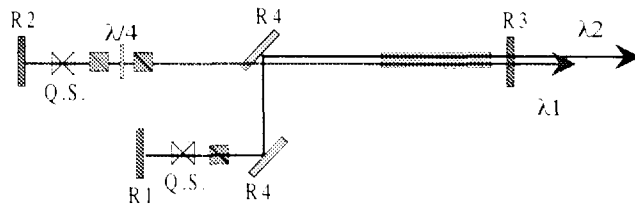


Fig. 3: Comparison of the performance of a hemispherical Nd:Cr:GSGG oscillator with a static mirror and with a variable radius mirror (VRM).

Nd:YAG lasing at 1064 nm has been one of the mainstays of the laser industry. Nd:YAG can, however, lase at other wavelengths. This offers a number of very interesting options for various applications while retaining all of the positive attributes of YAG. A program was undertaken to study the lasing action of Nd:YAG at 1319 nm not only in a single frequency oscillator but also in an oscillator simultaneously lasing at 1064 and 1319 nm. Figure 4 shows a schematic of the dual wavelength laser. Stable simultaneous lasing on two wavelengths using a single laser with substantially different stimulated emission cross-sections is a unique achievement.

Research into dual wavelength lasers started with the development of a model that allowed optimization of output energies, pulse durations, build-up times, and stable operation. For given output energies, the important parameters were found to be the dual wavelength output coupler reflectivities and the lengths of the lasing cavities. Also of key importance was the suppression of lasing on transitions other than those that gave the desired wavelengths. A design insensitive to changes in pump energy was found. Using this design, an oscillator that produced 100 mJ at 1319 nm in the single wavelength mode, simultaneously produced 60 mJ (1319 nm) and 100 mJ (1064 nm). Timing jitter (a parameter of importance in upconversion applications) was reduced to  $< \pm 1$  ns (5% of pulse width).

**Fig. 4: Schematic of the dual wavelength ( $\lambda_1=1319$  nm,  $\lambda_2=1064$  nm) Nd:YAG laser. R1, R2: High reflectivity (HR) mirrors @  $\lambda_2$  &  $\lambda_1$ ; R3: Output coupler with reflectivities  $R_a$  @  $\lambda_2$  &  $R_b$  @  $\lambda_1$ ; R4: HR @  $\lambda_2$  & high transmission @  $\lambda_1$ ; Q.S.: KD\*P Pockels cell Q-switch.**



### 2.3 Diode pumped lasers

Diode pumped solid-state lasers are attractive because: (a) their high average power performance far exceeds that of flashlamp pumped lasers, (b) they are more energy efficient, (c) they are more compact. Worldwide R&D and commercialization efforts in this field are extremely intense. We have chosen to concentrate on the development of high average power, high beam quality, continuously diode pumped lasers. The reasons are twofold: (i), the diode costs (\$/watt) are lowest for continuous diodes, and (ii), high average power lasers are the type required in medical and industrial applications.

In general, each generation of laser that we have built has surpassed its predecessor by an order of magnitude.<sup>(4,5,6)</sup> Our latest laser, the **Barak 10**, is a 10 W, CW and high repetition rate, short pulse laser with excellent beam quality.<sup>(6)</sup> It is being exhibited and marketed worldwide. Figure 5 presents typical performance curves for a **Barak 10**. Figure 6 shows a cross-section of the laser. The four diodes are close-coupled to a specially polished and coated Nd:YAG rod. The cylindrical surface of the rod acts as a focusing lens that concentrates the pump beam in an area that overlaps the low divergence mode of the Nd:YAG

laser that we generate. Both the diode and the laser rod are impingement cooled, i.e., water flows in channels that approach but do not directly contact the lasing elements. The YAG rod cooling geometry is restricted to the y direction in order to generate a one-dimensional Cartesian temperature gradient. This reduces birefringence losses in the Brewster face laser rod.

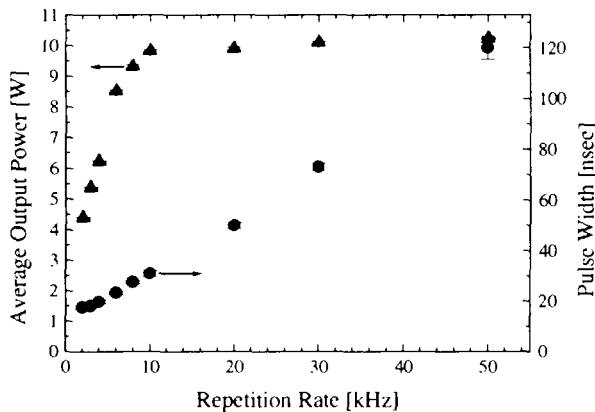


Fig. 5: Performance of the Barak 10 laser. The diode pump is continuous, whereas the Q-switch repetition rate may be varied over a wide range. The Q-switch may be turned off in order to obtain a 10 W CW beam.

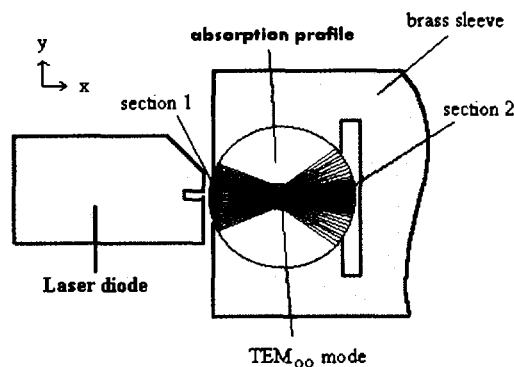


Fig. 6: Cross-section of the Barak 10 laser.

During the 1980s and 1990s, the use of potassium titanyl phosphate (KTP) became customary as techniques for growing large crystals (up to 2x2x4 cm) became common and as competition drastically reduced the cost. This crystal is particularly attractive for frequency doubling of 1000 nm light because the angular acceptance minimizes alignment and beam divergence requirements.

### 3. Frequency conversion devices

#### 3.1 Periodically poled KTP frequency doubling crystals

The crystal is widely used with pulse pumped (flashlamp and laser diode) Q-switched lasers that produce intensities of tens of MW in the crystal.

CW pumped Q-switched lasers generate lower intensities and therefore efficient frequency doubling is more difficult. Improved efficiency cannot be achieved solely by increasing the length of the crystals because, after a certain laser divergencedictated interaction length, the inverse down-conversion process starts. The onset of the inverse down-conversion is a result of accumulated phase mismatch between the fundamental and harmonic beams. If the phase mismatch could be eliminated, then longer crystal assemblies or more highly focused (higher divergence) beams could be employed.

Quasi-phase matching is a technique for controlling the phase mismatch between the fundamental and harmonic beams. After a certain amount of mismatch is allowed to develop, the crystal is altered so that the polarization matrix is reversed. The phase mismatch in this zone goes to zero, at which point the crystal is returned to its original state. This periodically structured crystal can have vastly superior frequency conversion properties compared with homogeneous bulk crystals.

The first crystal in which quasi-phase matching was attempted was lithium niobate ( $\text{LiNiO}_3$ ). Fundamental difficulties with the fabrication technique limited the depth of poling to only 300  $\mu\text{m}$ .

We obtained exceptionally good results by working with Periodically Poled KTP.<sup>(7)</sup> The Solid-State Physics group at Soreq, in coordination with a group from the



Electrical Engineering Faculty at Tel-Aviv University, successfully periodically poled 1-cm-long KTP crystals of high optical quality and superior nonlinear performance. In these crystals, a periodically varying voltage imprinted a periodic refractive index variation in the crystal. Using the *Barak 10* laser, we produced 6 W of green power in a high-repetition-rate Q-switched beam.

Other crystals with different poling periods were used in external cavity optical parametric oscillators to produce tunable mid-IR light.

### 3.2 Sum frequency mixing

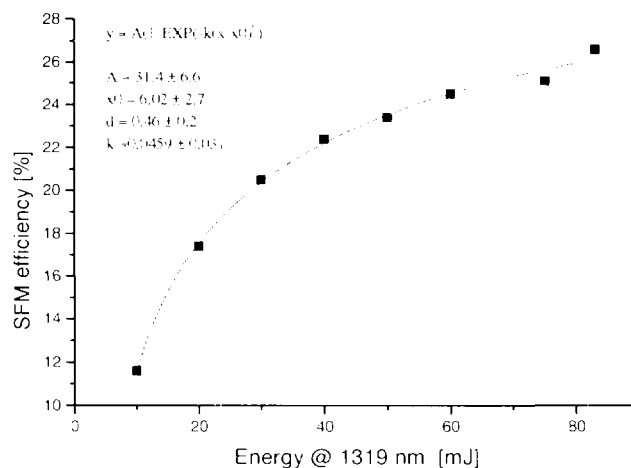
Frequency doubling is the process whereby two photons of the same frequency are combined into one, using the catalytic action of a nonlinear crystal. Over the past decade progress has been steady, with the conversion efficiencies of Q-switched pulse typically reaching 70% in KTP crystals.

Frequency tripling is another well known technique whereby the doubled output of one crystal is combined with the unconverted beam in a second crystal to generate a beam at triple the fundamental frequency. Typical tripling efficiencies in KTP are 40%. In fact, if an appropriate crystal is aligned in the correct orientation, any two photons can be added together to form a higher frequency photon. Sum frequency mixing is of interest to us because we have mastered the technique of locking together in time two short pulse lasers that generate photons at two different frequencies, as well as using Nd:YAG to generate pulses simultaneously at two frequencies. Using Nd:YAG, we can

generate photons at 1064 and 1319 nm. Doubling each laser separately generates 532 nm (green) and 660 nm (red). Sum frequency mixing the two fundamental wavelengths yields 589 nm (yellow). Thus, we have generated five separate wavelengths using a single lasing material.

Using two separate Nd:YAG lasers, each lasing at separate wavelengths, we obtained 60 mJ of yellow light at an efficiency of nearly 30% (Fig. 7). Using a single laser emitting two wavelengths, we obtained 20 mJ of yellow light at an efficiency of 15%. Performance was limited by available input energy at 1319 nm.

**Fig. 7: Upconversion efficiency for the sum frequency mixing (SFM) of 1064 and 1319 nm synchronized pulses.**

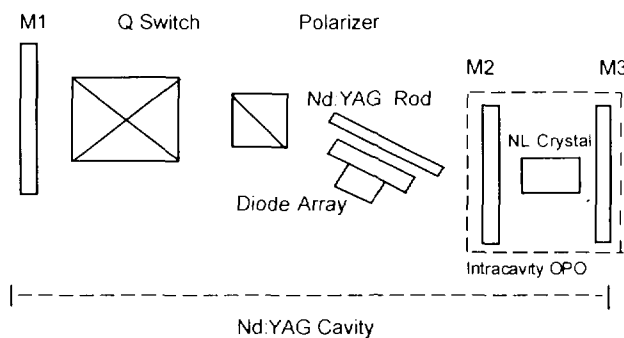


### 3.3 Optical parametric oscillators

Lasers are excellent sources of coherent light. They are, however, limited to emitting at specific frequencies or to confined spectral bands. Optical Parametric Oscillators (OPOs) use difference frequency mixing to obtain continuously tunable coherent light over extremely broad spectral ranges. For instance, one of our first OPOs used frequency tripled light from a Nd:YAG laser and was continuously tunable throughout the entire visible portion of the spectrum and well into the near infrared.<sup>(8)</sup>

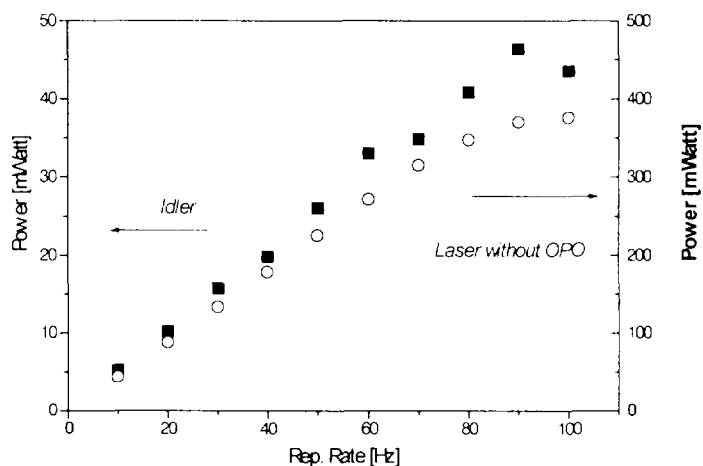
Difference frequency mixing (the inverse of sum frequency mixing) is the process by which a single photon is split into two lower frequency photons. The efficiency of this process is enhanced, when the build-up starts from noise, by building a resonator (two mirrors that reflect one or both of the difference frequency photons) around the nonlinear mixing crystal. The resulting device is an optical parametric oscillator (OPO).

Because of the importance of 3-5 $\mu\text{m}$  tunable light sources for applications such as remote sensing, we have devoted considerable effort to the development of mid-IR OPOs. Prior to the invention of periodically poled crystals, we built extra cavity OPOs for attachment to high energy lasers and intra-cavity OPOs for incorporation into lower power, high repetition rate Q-switched oscillators.<sup>(9)</sup> Figure 8a and 8b shows an example of the intracavity laser and its mid-IR performance.



**Fig. 8a: MID-IR intracavity optical parametric oscillator (OPO) imbedded in a diode-pumped laser.**

**Fig. 8b: Intracavity optical parametric oscillator (OPO) output compared to standard laser output**

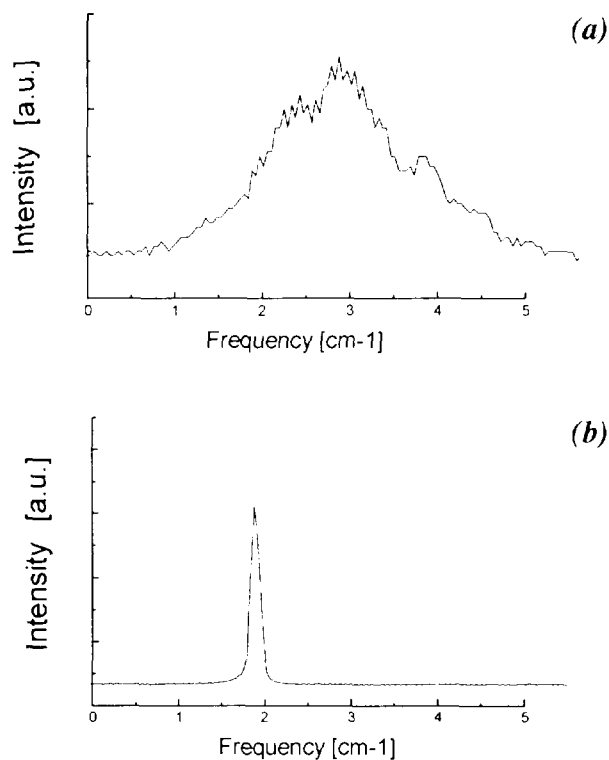


### 3.4 Narrow linewidth OPOs

Narrow line width infrared OPOs are required for remote sensing lidar systems. Linewidth can be narrowed by injection locking the OPO, i.e., injecting a narrow bandwidth seed pulse or by inserting a linewidth narrowing grating into the OPO cavity and tuning it to the appropriate wavelength. One problem of the first approach is the need to find a laser source that emits at the proper infrared wavelength; the second approach has the disadvantage of increasing the losses within the OPO and substantially reducing the OPO output.

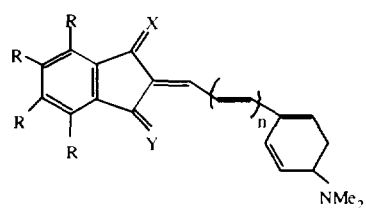
We have devised a hybrid solution that takes advantage of the OPO's properties.<sup>(10,11)</sup> The pump laser is narrow bandwidth; therefore, if one of the two OPO frequencies is narrow linewidth, then the second frequency will also be narrow linewidth. If a dye laser is tuned to the short wavelength OPO frequency (signal), then the infrared output (idler) will also have the desired bandwidth. The dye laser and the OPO can be combined into a single coupled cavity. Narrowing the linewidth of a dye laser is easy because the high gain of standard dye lasers compensates for the extra losses of the spectral grating. The end result is

a compact, efficient, and robust narrow-linewidth ( $0.2 \text{ cm}^{-1}$ ) source (Fig. 9).



**Fig. 9: Bandwidth of unseeded (a) and seeded (b) optical parametric oscillator**

Our activities are aimed at the development of new, thermally stable and highly efficient nonlinear optical (NLO) chromophores, as well as electro-optic (EO) and photorefractive (PR) polymers. These materials could be used in electro-optic integrated optical devices for ultra-fast light modulation (EO polymers) and dynamic memories (PR polymers). Various promising chromophores with 1,3-indandione-2-ylidene moiety as an efficient electron accepting group within donor-acceptor push-pull polyenes, have been synthesized and studied.



## 4. Nonlinear optical materials development

### 4.1 Second order nonlinear chromophores and polymers

The electronic structure of these 1,3-diketone derivatives can be tuned easily by the further chemical modifications that makes these compounds attractive for detailed investigations of 'structure - properties' relationships. Second-order nonlinear properties of the chromophores are studied by EFISH (electric-field-induced SHG), and by Hyper Rayleigh Scattering (HRS) at 1.06 $\mu\text{m}$  and 1.54 $\mu\text{m}$ . The results of  $\mu\beta$  obtained by EFISH for various new chromophores are presented below.<sup>(12)</sup>

**Table 1 - Results of the electric field induced second harmonic measurements.**

Compound	$\lambda_{\text{max}}$ (nm)	$\mu\beta_0 \times 10^{-48}$ esu	
1	534	230*	1) R=H, X=Y=O, n=1
1(NEt <sub>2</sub> derivative)	552	399*	
2	570	500*	2) R=F, X=Y=O, n=1
3	592	966* ; 460**	3) R=H, X=C(CN) <sub>2</sub> , Y=O, n=1
4	670	>1000*	4) R=H, X=Y=C(CN) <sub>2</sub> , n=1
5	604	280**	5) R=H, X=Y=C(CN) <sub>2</sub> , n=0

\* In Chloroform;

\*\* In Toluene

Electro-optic polymers were prepared from the new chromophores as guest-host and sidegroups in optical polymers. Corona poling techniques have been used and thoroughly studied (high temperature poling under inert conditions for PVK polymers). Evaluation of the EO polymer properties is carried out by SHG from thin poled films.

We have also studied a unique photochromic dye with large second-order nonlinearity. The transition is based on a reversible formation - cleavage of a C-C bond. The bicyclic di-indandione derivative ( $\lambda_{\text{max}}=480\text{nm}$  in toluene)

undergoes a reversible photochemical ring opening to form an isomer ( $\lambda_{\max}=640\text{nm}$  in toluene). This isomer constitutes a conjugated donor-acceptor system and exhibits considerable second-order optical nonlinearity as found by electric field induced SHG (EFISHG) measurements. The photochromic conversion is observed also in the crystalline form indicated visually by crystal color change from red to green.

Nonlinear asymmetric zeolites, which were doped with chromophores within the pores but which maintained their polar ordering, were studied using the Kurtz powder technique and two-photon absorption. High frequency doubling efficiencies were observed ( $> \times 10$  quartz). New organic salt crystals (quinoline derivatives) which possess polar order were also studied and found to show large NLO activity ( $> 2x$  urea) in powder tests. These crystals are UV-transparent.

The microsphere activity has developed around several different aspects, corresponding to different physical effects and microsphere sizes. Typically, the activity can be divided into two main subjects: (i) micromanipulation and testing of nonlinear optical microspheres, and (ii) laser action in diffusive media.

This project is devoted to the development, study and manipulation of microspheres with giant nonlinear optical properties. These properties are due to degenerate spherical optical modes of the microspheres giving rise to giant optical resonance (morphology dependent resonance

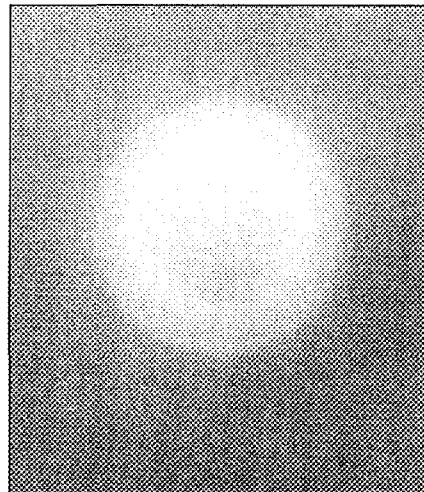
## 4.2 *Optical properties of microspheres*

### 4.2.1 Nonlinear optical microspheres

MDRs). Most of the study has been dedicated to the microlaser.

Microlasers consist of polystyrene microspheres filled with DCM laser dye. After a careful choice of high sphericity microspheres ( $\sim 40\mu\text{m}$  diameter), we obtained lasing from the microspheres by pumping directly with green, pulsed laser light focused by a microscope. A typical magnified image of a lasing microsphere is shown in Figure 10. Because the MDRs are circular modes propagating near the surface, one obtains a strong light enhancement near the surface.

In order to study the coupling of lasing microspheres with different micro-optical components, we have built an optical tweezers system which allows precise 3D manipulation of the microspheres. The next stage is thus to study the coupling efficiency of a lasing microsphere with an optical waveguide and maximize this coupling for future micrometer-sized electro-optical devices.



*Fig. 10: Magnified image of a lasing microsphere.*



Following the recent discovery of laser action from diffusive media containing laser dyes, we initiated a series of experiments aimed at characterizing and optimizing these effects. The applications of such inexpensive lasing media cover the range from medical applications (especially skin treatments) to product identification (in warehouse and supermarket inventories).

Dye-methanol solutions containing suspensions of 32 nm diameter  $\text{TiO}_2$  (titanate) particles exhibited lasing action when excited with a pulsed, frequency doubled Nd:YAG laser (532 nm). When the solution did not contain scatterers, increases in pump power resulted only in an increase in the dye luminescence (limited by dye saturation). However, when the concentration of the scatterers increased, the emission did not saturate with increasing pump power, as occurred with luminescence, but increased linearly. Moreover, one could observe a measurable reduction of the emission linewidth, from tens of nanometers down to less than 5 nm, as well as an increase in the light emission efficiency. Similar results have been obtained in dye-doped plastic films, showing the flexibility of this class of materials.<sup>(13)</sup>

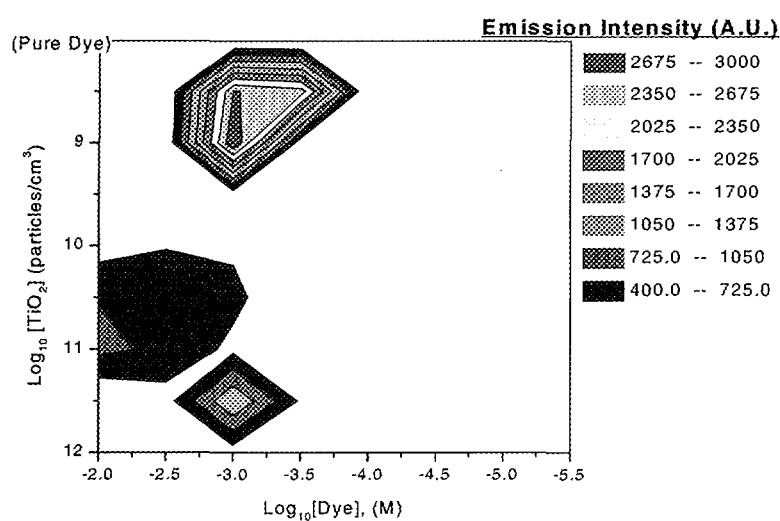
After obtaining the preliminary results that showed lasing-like action in dispersions of titanate nanospheres in laser-dye solution, and the experimental evidence of optimum conditions for lasing, our efforts were concentrated on elucidation of the mechanisms which caused the effect, in order to further optimize such lasing. Therefore, several experimental techniques as well as a simple theoretical model were developed.

#### 4.2.2 Laser action in diffusive media

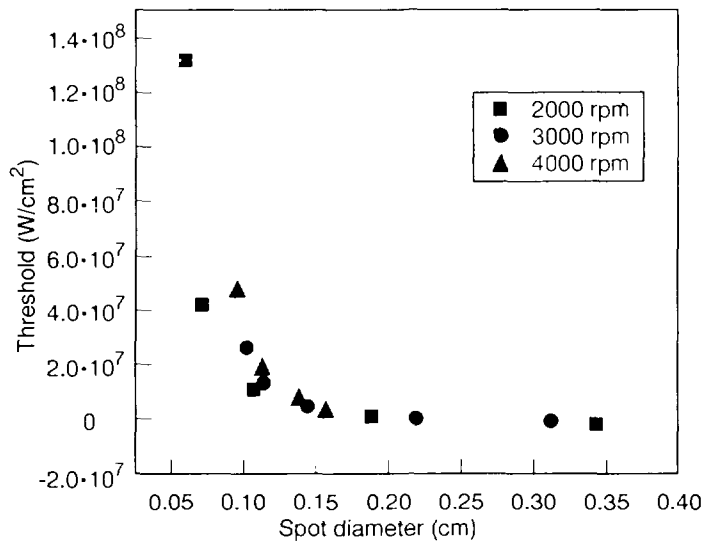
## 4.2.2.1 Mapping techniques

A series of systematic experiments were conducted in which the dye concentration and the scatterer density were varied independently, and we investigated how these parameters affected the lasing process. It was discovered experimentally that there exist completely isolated lasing regimes. Figure 11 (experimental data for high pump energy) depicts these regions. Typically, one can see three lasing zones, one at very high dye (rhodamine 6B) concentration, and two around  $10^{-3}$  M dye concentration. The high dye concentration lasing zone is not relevant to the problem at hand and corresponds to strong chemical interaction between titanate particles negatively charged and rhodamine ions. However, two different regimes appear clearly at low and at high titanate densities for roughly the same dye concentration. The low- $\text{TiO}_2$ -density enhancement is due to weak scattering of amplified spontaneous emission towards the detector, whereas the high- $\text{TiO}_2$ -density enhancement is due to trapping of photons inside the highly scattering gain medium.

Fig. 11: Parametric mapping of diffuse media lasing



Once it was established that a lasing effect can result from photon trapping due to high scattering, it seemed clear that the threshold for lasing had to depend on the pump spot size. Experiments with thin dyed polymeric films filled with titanate nanospheres showed directly that the sample macroscopic size influenced the lasing threshold.<sup>(14)</sup> Three different samples with thicknesses varying between 1 and 10 $\mu\text{m}$  were used in deriving the results presented in Figure 12. It can be seen that the threshold is independent of the thickness. However, the threshold appears to vary with  $L^{-3.2}$  (where L is the focal spot diameter).



#### 4.2.2.2 Spot size dependence threshold

*Fig. 12: Dependence of threshold on pump focal spot size for lasing in the trapped photon mode.*

With our system the lasing threshold ( $P_{th}$ ) can be determined directly and precisely. Since the lasing threshold is a basic parameter in lasing theory, investigation of the effect of various parameters on the threshold enables comparison between experiment and theory and can be expected, therefore, to lead to further understanding of the lasing phenomena in diffusive media.

#### 4.2.2.3 Photon diffusion model

Using a combination of laser and photon diffusion theory, we were able to derive analytical expressions for critical parameters such as the threshold. Thus for the lasing threshold as a function of pump fluence the following expression was derived:

$$P_{th} = 4\pi h\nu l^* / L\sigma\tau_f\eta_Q \quad (1)$$

where  $h\nu$  is the photon energy,  $l^*$  is the photon transport length, and  $\sigma\tau_f\eta_Q$  are specific parameters of the lasing material (dye in our case). Although reasonable agreement was obtained between the model and the experimental result both the experiment and the theory still need improvement in order to better understand the physical mechanisms.

### **4.3 Materials with efficient nonlinear absorption**

We have studied materials which possess optical limiting capabilities due to their nonlinear absorption properties. Such mechanisms are known to be fast (subnanosecond response time) and do not exhibit increased scattering (unlike suspensions of absorbing particles). Since two photon absorption is an intensity dependent process, and since known materials typically have high intensity threshold, we have directed our attention to another class of materials: those which exhibit reverse saturable absorption (RSA) properties. This optical limiting mechanism is fluence dependent and rapid (on a picosecond time scale). Strong limiting effects based on RSA have already been demonstrated in several organic molecules. The RSA mechanism results from excited state absorption with a stronger absorption cross-section than the ground state (at certain desirable frequencies). The excited state is either an excited singlet ( $S_1$ ) state or a triplet ( $T_1$ ) state populated by

an efficient intersystem crossing mechanism. The  $S \rightarrow T$  transition is the main mechanism responsible for RSA in metallo-phthalocyanines (M-Pc) and metallo-porphyrines (M-Por). We have studied the RSA properties of heavy metal phthalocyanines, starting with commercial products. Commercial Pb-Pc was purified to yield two fractions:  $Pb^{+4}$ -Pc and  $Pb^{+2}$ -Pc. The former fraction exhibited a much stronger nonlinear effect than the latter. It was also found out that oxygen in the solution induces strong quenching effects. The quenching almost eliminates the nonlinear effect unless special care is taken during solution preparation. This is particularly true for the highly efficient RSA material Pb-Pc, whereas it was found to play no role for Zn-Pc. Z-scan measurements carried out in toluene solution with a 10ns pulsed laser at 532nm yielded the absorption cross-sections shown in Table 2 (fit based on a model assuming efficient and fast  $S \rightarrow T$  transition, and long-lived T state).

M-Pc	$\sigma_{01}$	$\sigma_{34}$	$\sigma_{34} / \sigma_{01}$
$Pb^{+4}$ -Pc	2.75	188	68
$Pb^{+2}$ -Pc	4.75	18	3.8
$Zn^{+2}$ -Pc		98	

*Table 2 - Absorption cross-sections measured for reverse saturable absorbers (RSA).*

As can be seen from Table 2, the reduced fraction of Pb-Pc has a much lower nonlinearity. The sensitivity to oxidation state of the molecule led us to investigate of the effect of oxidation. For this purpose we recently initiated electrochemical studies of Zn-Pc which is not sensitive to the presence of oxygen. A specially designed cell, suitable for electrophotochemical investigation, is in preparation and

will enable investigation of oxygen-sensitive compounds such as Pb-Pc. Solid samples of sol-gel containing Zn-Pc have also been prepared. Specially designed metallo-Pc compounds are currently being synthesized. These new materials will enable us to elucidate the role of side groups in RSA activity as we change from strong donors to strong acceptors.

## **5. Remote sensing**

Laser-based remote sensing systems are ideal for detection and analysis of air, water, and land-surface borne chemicals, because there is no need for close physical contact between the sensor / operator and the unknown agents, and because the range and field of view of the detection system may be large. We have concentrated on the development of Differential absorption lidar (DIAL) techniques. In this technique, one looks at the absorption spectra of the target molecules. By analyzing two frequencies per absorption feature, one at the absorption peak and one just beyond the absorption line (looking at the background), signal variations due to extraneous sources can be factored out and the system sensitivity can be increased substantially.

Many chemicals have strong absorption features in the infrared. For this reason our IR OPO development has found an ideal application. A DIAL OPO must not only be tunable to the right frequency, but must also be frequency stable and must produce photons on and off the absorption line. Our work has shown that narrow linewidth OPOs are not suited to the requirements of DIAL systems. Instead, it was found that OPOs with a spectral width that extended

several times beyond the width of the targeted absorption line had the required characteristics of frequency stability and on / off line photon production, as well as simplicity of design.

To use this wide bandwidth DIAL transmitter required the development of a special detection technique.<sup>(15)</sup> Soreq is the first to utilize this gas-correlation receiver (GCR) in an active DIAL system. Simply put, the GCR splits the return signal into two branches: one part goes directly to a detector and gives a measure of the total return signal; the other part goes through a cell containing a sample of the chemical being sought, and then to a second detector. If there were traces of the chemical in the atmosphere, then the absorption feature would have reduced the on-line light intensity even before the gas cell was reached. The extra absorption of the gas cell would have a relatively small effect. If there were no traces of the chemical in the atmosphere, then the gas cell would have a relatively greater effect. This procedure can increase the sensitivity one hundred fold over that obtained with conventional DIAL systems.

Optics is usually considered as useful for devices larger than the wavelength of the light. This is due to the fact that the interaction of light with sub-wavelength features, in any medium, leads to energy loss through decay in the form of evanescent waves. However, in recent years great strides have been made in the development of microscopes (near-field scanning optical microscope, or NSOM) which break this diffraction barrier. Light which is forced through sub-wavelength apertures can sample and

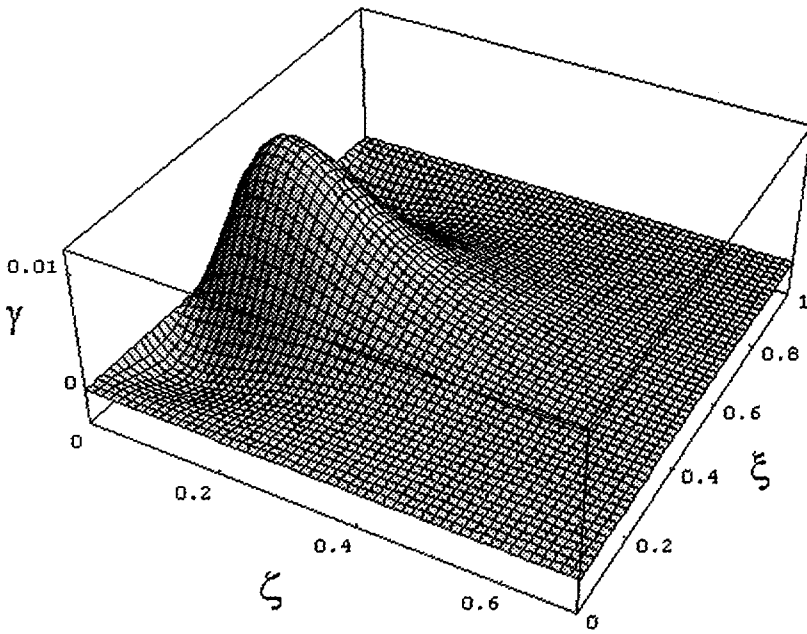
## ***6. Nonlinear optics and light-matter interaction in the near-field regime***

acquire information concerning sub-wavelength spatial features in media which are in the 'near-field' of the light, i.e., at distances smaller than the aperture's diameter. In this fashion, images are acquired with a resolution much higher than the diffraction limit would normally allow.

The Nano-Optics Laboratory at Soreq NRC is dedicated to the theoretical and experimental study of nonlinear and other types of light-matter interactions in the sub-wavelength spatial regime. Applications to be developed include high-density optical memories, nano-sized optical devices for integrated optics, and improvement of the NSOM technology for imaging.

A theoretical study of the interaction of light exiting a sub-wavelength aperture with Kerr medium (where the refractive index  $n$  depends on the light intensity  $I$  via  $n=n_0 + n_2 \cdot I$ ), leads to new and unexpected predictions concerning the light's response.<sup>(16)</sup> Due to the strong spatial gradients in the light intensity, it is possible to induce convergence of the light through interaction with a negative Kerr medium. Figure 13 displays this 'light-bunching' effect. We are currently studying other configurations in order to optimize and possibly create self-channeling of the sub-wavelength light. This would have important applications, for example in forming sub-wavelength wave guides and in guiding light through sub-wavelength apertures with improved efficiency.





*Fig. 13: Self-convergence of light with sub-wavelength spatial dimensions.*

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