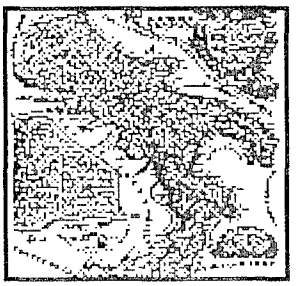
5th Mediterranean Workshop and Topical Meeting

"Novel Optical Materials and Applications"

NOMA '01



ABSTRACTS

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14. ABSTRACT

The Final Proceedings for 5th Mediterranean Workshop on Novel Materials and Applications (NOMA), 20 May 2001 - 27 May 2001

This is an interdisciplinary conference. Topics include novel optical materials like semiconductors, polymers, liquid crystals, composite materials, photorefractive and electro-optical crystals and reduction to practice in actual devices.

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ABSTRACTS

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Nonlinear quantum magneto-optic micro-cavities

and photoinduced spin dynamics

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Polarization state sensitive effects and their relation to fundamental aspects of the nonlinear light matter interaction are attracting much attention as they are relevant to important applications as well with some far reaching repercussions. This is particularly striking when non reciprocity comes into play as in the case of magneto-optical interactions in magnetic systems in confined geometries. In the case of strong charge photon spin coupling regime in quantum confined nano-structures they can appreciably affect the spin organization and dynamics and provide ways of unidirectional control of their coherence and transport.

We shall present recent studies of the nonlinear quantum magneto-optic micro-cavities containing semi-magnetic semiconductor nano-structures and show that strong charge-photon-spin coupling has a polarization specific impact on the transmission characteristics and exciton-polariton behavior. We will also review and discuss the recent work on photo-induced Faraday rotation and use it to study spin reorganization and dynamics in such semi-magnetic semiconductors and other hetero-structures and interfaces.

Picosecond Reverse Saturable Absorption and Optical Limiting in Molecular Materials, Fullerenes and their Metal Derivatives

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Abstract

Investigations of the reverse saturable absorption behaviour of Porphyrins, soluble Phthalocyanines, Fullerenes C60 and C70 and some of their Pt and Pd metal derivatives have been carried out with picosecond pulses at 532nm. From intensity dependent transmission measurements, coupled with a population level kinetic analysis, excited state cross sections were determined for the materials. Of the Fullerenes studied here, C60 offers the most efficient limiting ability but it appears to be less attractive than some heavy metal phthalocyanines at 532nm. It can be seen that the transmission drops to half its low level value at an intensity of ~3 GW/cm2. For 35ps excitation this correspond to a fluence of 0.1J/cm2. This is 105 times higher than the maximum permissible exposure for retina damage. Wavelength dependent studies show that the sex/s0 ratio increases rapidly from 532nm to 680nm and is larger than that of CAP or SiNc. The triplet state absorbs strongly in this region. However, the ground state absorption declines rapidly after 620nm so it is unlikely that it would be any more efficient at wavelengths greater than this than at 532nm but it may offer improved performance up to 620nm. C70, due to its large ground state absorption, has been shown not to be an efficient limiter. The metal derivatives perform better than C70 but less efficiently than C60.

PLASMONIC MESO- AND NANO-STRUCTURES: NEW AVENUES FOR PHOTONICS, LASER PHYSICS AND SPECTROSCOPY

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Fundamentals of electromagnetic and optical properties of micro- and nano-structured metal-dielectric composites, both ordered and disordered, are reviewed. We analyze how symmetries of different plasmonic materials influence their optical properties [1,2]. By employing unique properties of metal nanostructures, we show a possibility of the fabricating of low-loss plasmonic crystals with a large and scaleable photonic band gaps. We also show that in periodic metal mesostructures, the extremely low-frequency plasmons can be excited. The periodicity makes possible sneaking light through sub-wavelength hole arrays, leading to extraordinary optical transmission, which can be controlled by light itself. By combining periodic and percolation nanocomposites one can design left-handed "metamaterials" with a negative refractive index, which have unique optical properties and can act, for example, as perfect lenses. Finally, the scale-invariant fractal symmetry of disordered nanocomposites results in Anderson localization of light by nanometer-sized plasmonic resonators, where the local field exceeds the applied field by many orders of magnitude and optical nonlinearities are dramatically enhanced [1,2]. The electromagnetic modes focused within nm-sized "hot spots," acting like nano-antennas, make possible a number of novel applications, such as low-threshold microlasers, super-dense optical recording, sub-fs pulse generation, Raman and nonlinear spectroscopy of single molecules and quantum dots, enhanced photochemistry and photobiology.

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Laser-induced ripple structure of dopant on the substrates in a dye-doped liquid crystal cell and its alignment effect

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Abstract

Photo-induced reorientation effects in dye-doped liquid crystals (DDLCs) have received considerable interest recently. Related studies have indicated that several mechanisms contribute to such reorientation effects [1-5]. Janossy et al. [1] observed that photo-excited anthraquinone dye molecules induced a positive torque, thereby facilating the reduction of the optical Freedericksz transition. Gibbons et al. [2,3] demonstrated that nematic liquid crystals were reoriented perpendicularly to the optical electric field, if the substrate coated with an azo-dye doped polyimide was excited with a linearly polarized light. In other words, the photo-excited azo dyes induce a negative torque in this case. While observing another dye-induced reorientation effect, Khoo et al. [4,5] indicated that the dye-induced space charge field could induce the reorientation of the nematic liquid crystals.

In general, a doping dye could result in one of the above orientational effects. However, some dyes could induce several effects simultaneously. Among these effects one may dominate, depending on the experimental conditions. For example, methyl red (MR) dyes, after photo-excitation, could develop trans-cis isomerization [6], surface alignment effect [7,8], and photorefractive-like effect [4]. During the dynamic measurements, the trans-cis and photorefractive-like effects dominate in the early time, while the surface alignment effect dominates in the later stage.

In this talk, we report on the formation of laser-induced ripple structure of the adsorbed MR dyes on substrate surfaces in a DDLC homeotropical cell. Permanent reorientation of the liquid crystal molecules is accompanied with the formation of the ripple pattern. To our knowledge, this study reports on such an effect for the first time. Previous studies have observed laser-induced periodic surface structures (LIPSS) on the surface of metals [9,10], semiconductors [11,12], and dielectrics [13,14], using either the CW or pulsed lasers with the wavelengths ranging from 0.53□ m to 10.6□ m. Explanations of LIPSS effect were given in terms of the coupling of laser-induced acoustic modes [15], or the driven surface plasmons [16]. Microroughness is crucial in the latter model.

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Novel Crystallization Method, Ring-heater Heated Pedestal Growth Method, for Nonlinear Optical Organic Material

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Abstract

Organic nonlinear optical (NLO) materials are good candidates as materials for signal processing device used in the high-speed optical communication system. However, difficulty to grow large single crystals toward a required direction, i.e., a phase-matched direction is their critical issue for real applications. Recently, variety of approaches for the issue has been investigated. For example, one of the melt growth techniques, indirect laser-heated pedestal growth (ILHPG) method had been developed to control the crystal growth. However, this method has the problem that laser heating system is high in cost and complicated. Here, we will report a novel and simple crystal growth method, ring-heater heated pedestal growth (RHHPG) method that made it possible to control the growth direction toward a phase-matched direction. As an organic NLO material, we used a 2-damntylamino-5-niteopyridine (AANP), which has large NLO properties d_{33} of 80 pm/V. Melting point of AANP is 167 °C. Schematic diagram of RHHPG method is shown in Fig. 1. We used a ring-heater instead of the CO_2 laser and focusing system in ILHPG method to melt the source materials. Figure 2 shows the vertical temperature gradient of the heating along with that of ILHPG method. The molten zone temperature distribution by RHHPG was almost the same as that of ILHPG. We tried to obtain the crystal with growth direction controlled toward a phase-matched direction. Phase-matchable crystals at 1.3μm and 1.55μm wavelengths were obtained by this method.

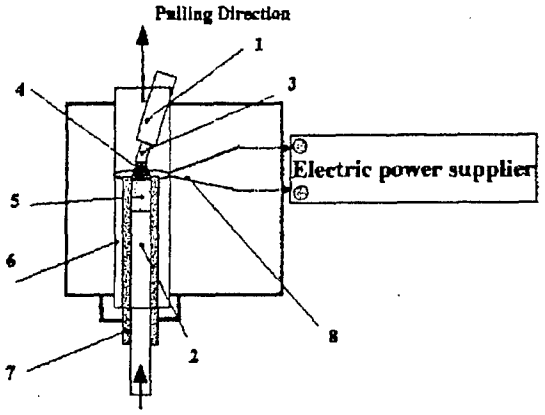

Fig. 1 Schematic diagram of RHHPG.
1: Pulling rod; 2: Pushing rod; 3: Seed crystal;
4: Molten zone; 5: Materials; 6: Inner glass tube;
7: Teflon tube guide; 8: Ring-heater

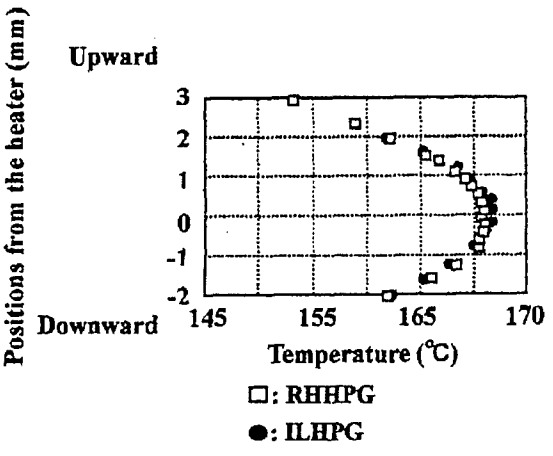

Fig. 2 Vertical temperature gradients in the molten zone.

FREQUENCY CONVERSION IN PERIODICALLY POLED LITHIUM NIOBATE WAVEGUIDES

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The cascade of two second-order processes in a nonlinear optical crystal can be used to give rise to a third-order process, such as nearly-degenerate four-wave mixing. The approach is attractive because the effective third-order nonlinearity produced by cascading can be not only large, but also fast and non-absorbitive. Various experiments have been performed in the past few years by using both inorganic and organic crystals, showing that wavelength converters potentially interesting for optical communications could be realized by using such an approach. The challenge is now to obtain an efficient conversion in a waveguided device. The talk will focus, in particular, on experiments performed with channel waveguides obtained on a periodically poled lithium niobate substrate. It will be shown that it is very important to perform an accurate modal analysis of the nonlinear interaction, and that very severe requirements might arise on the waveguide uniformity.

TWO-PHOTON 3D MICROFABRICATION OF POLYMERS AND METALS

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ABSTRACT

Recent progress in the development of materials for 3D microfabrication of polymeric and metallic structures by two-photon direct laser writing will be presented.

SUMMARY

3D microfabrication of polymeric and metallic structures by two-photon laser scanning direct writing is of interest for integrated and micro-optics, MEMS, microfluidics, and other applications. High sensitivity two-photon radical and acid generating initiators which can be used to polymerize acrylates and epoxides have been developed. Microfabrication of polymer microstructures in these systems, as well as in chemically amplified resists will be described. We will also describe a new approach to the direct writing of 3D metallic structures within a metal nanoparticle/polymer composite. Related composites have also been patterned using electron beam exposure offering potential direct e-beam lithography of metal features. Prospects for applications, the extension of this approach into the nanoscale, and for the preparation of hierarchically structured materials will be discussed. Recent results on the enhancement of two-photon absorption of chromophores near clusters of metal nanoparticles will also be discussed. Enhancements by factors of up to 10,000 are observed for two-photon dyes on clusters of silver nanoparticles. The enhancements are spatially inhomogenous and frequency selective, in agreement with theoretical predictions. This surface enhancement of two-photon absorption offers interesting possibilities for sensing and imaging applications.

Design and Applications of Molecules with Large Two-Photon Absorption Cross Sections

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We have developed structure-property relationships for two-photon absorbing conjugated organic materials and on the understanding of the underlying physical mechanisms. As a result of this effort, a strategy for the design of molecules with large two-photon absorption cross sections, δ, has been developed. The strategy is based on the concept that symmetric charge transfer upon excitation, from the ends to the middle (or vice versa) of a properly substituted conjugated molecule is correlated with increased δ. This has led to the synthesis of molecules with δ up to 20 times larger than previously available chromophores. For example, 4,4'-bis(di-*n*-butyl)amino-*E*-stilbene exhibits a value of 210×10^{-50} cm^4 sec/photon at 605 nm, which is almost 20 times larger than that of *trans*-stilbene.

Two-photon excitation can be used to activate a variety of chemical and physical processes with high 3D spatial resolution. The electron richness of 4,4'-bis(di-*n*-butyl)amino-*E*-stilbene and related molecules has been exploited to activate two-photon induced polymerization of acrylate monomers with high efficiency [2]. While these molecules are much more efficient two-photon initiators than conventional dyes designed for one photon initiation, they had not been optimized for their chemical initiation efficiency. We will describe approaches to improve the sensitivity of our two-photon dyes for the generation of reactive species. In addition, we will describe preliminary studies on the use of two-photon fluorescent dyes for biological imaging applications.

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Vector Optical-Phase Conjugation in ZnS

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One striking feature of phase-conjugate mirrors (PCMs) is the handness-conservation: both incident and reflected light maintain the same ellipticity, in contrast with conventional mirrors that reverse the handness of elliptically polarized beams. Of course, any anisotropy of the PCM will affect the quality of the polarization conjugation and there is currently great interest in searching for a polarization insensitive scheme, primarily motivated by the possible use of phase-conjugation in all-optical fibre communication networks.

In this work, we present our experimental results on the polarization properties of the light reflected at a semiconductor PCM. Optical phase-conjugation was realised by degenerate four-wave mixing (DFWM) in ZnS by pumping the crystal with 100 femtosecond pulses below half the bandgap ($\_\_{}_0 \approx 0.43E_g$). The pump beams were co-linearly polarized, while the polarization state of the probe beam was changed continuously from co-linear (with respect to the pumps) to almost circular. The polarization of both the probe and the signal (conjugated) beams were measured by the polarization compensation method and the results have been analysed in terms of polarization azimuth angle α and ellipticity ε. In Fig.1*a, b* we show the experimental results (circles) together with the outcome of a simple transfer-matrix calculation (solid lines) for our specific set-up. Dotted lines labelled *1* show the behaviour of conventional mirrors ($\alpha_s = -\alpha_p$, $\varepsilon_s = -\varepsilon_p$) and dotted lines labelled *2* show the expected behaviour of an ideal, perfectly isotropic PCM ($\alpha_s = -\alpha_p$, $\varepsilon_s = \varepsilon_p$).

ZnS is a cubic semiconductor whose third order susceptibility tensor has non-zero off-diagonal terms. With co-linearly polarised pumps, the orthogonal polarization is excited by $\chi^{(3)}_{xyyx}$ term, whose value, in the limit of long wavelengths, is 1/3 of the diagonal term $\chi^{(3)}_{xxxx}$, effective when both pumps and probe have the same polarization. We measured the ratio of the conjugate-reflection coefficients for the intensity of linearly polarized light parallel and perpendicular to the pump polarization ($R_{\_}/R_{\_} = 12\pm4$), and used this value to calculate the solid curves in Fig.1. The strong nonlinear behaviour of the ZnS-PCM is very well described by our model calculation, which takes into account the experimentally measured linear polarization behaviour of the optics and the coating of the sample, without any fitting parameters. The small deviations from the experimental data are possibly due to the nonlinear-induced birefringence of ZnS whose effect is not included the present model. Our ZnS-PCM clearly preserves the sign of the ellipticity (shaded regions in Fig.1*b*), even though it is not polarization insensitive. We are currently exploring different pumping schemes in order to improve the polarization response of the ZnS-PCM.

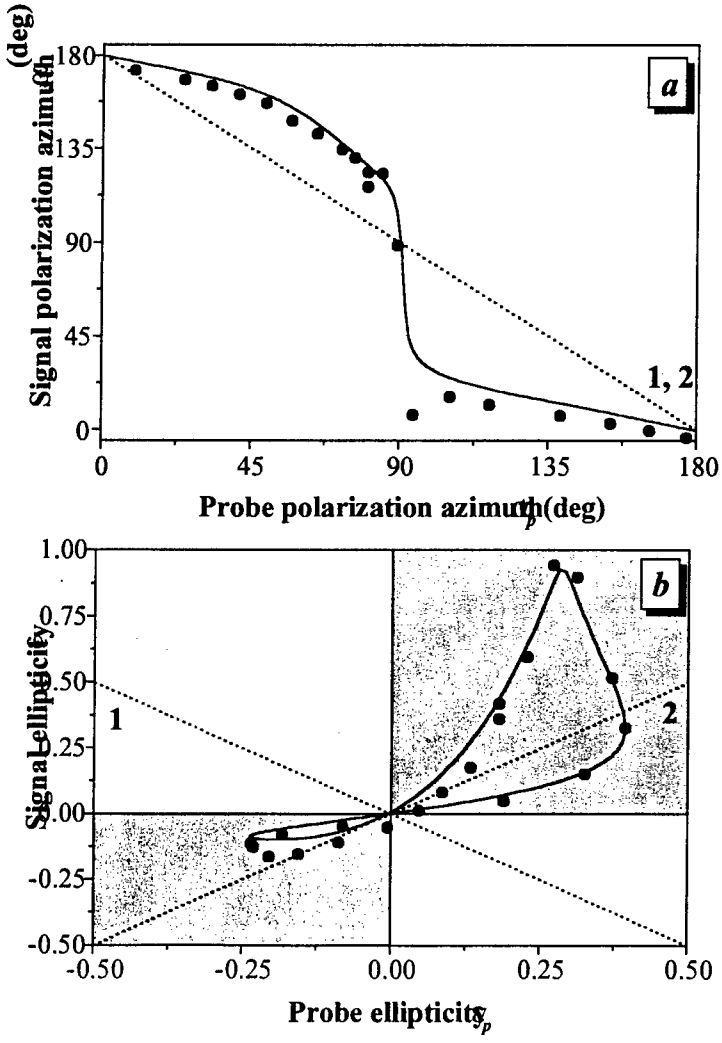

Fig.1. Polarization azimuth (*a*) and ellipticity (*b*) of the conjugated (signal) beam versus the same parameters of the probe beam. Circles are experimental data and solid lines are theoretical calculation. Dotted lines 1, 2 show the behaviour of ideal conventional and polarization insensitive phase-conjugate mirrors, respectively.

Third-order optical nonlinearity enhancement of gold nanoparticles embedded in a dielectric host

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Noble metal nanoparticles embedded in a dielectric host exhibit large optical nonlinearities [1], making these materials promising candidates for the realization of photonics devices for optical communications. The nonlinearity enhancement is due to the local electric field which is highly amplified at the surface plasmon resonance (SPR).

We present a study of Au:SiO_2 samples prepared by a sputtering technique. Several characterization methods have provided information about the sample morphology and the linear optical properties. The real and imaginary parts of the 3rd order nonlinear susceptibility $\chi^{(3)}$ have been measured in the nanosecond regime by z-scan experiments [2]. Results have revealed a large negative $\mathrm{Im}\,\chi^{(3)}$ value, at the SPR, increasing with the metal concentration. This increase is larger than the one predicted for low metal concentrations, and is thought to be due to interactions between particles.

At the same time, local electromagnetic field calculations have been performed by means of a recursive transfer matrix method recently developed [3]. The variations of the local electric field inside the metallic clusters have been studied as a function of the applied field frequency and the distance between particles. These calculations give a better understanding of the experimental results for $\chi^{(3)}$.

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MICROSTRUCTURED POLYMER BASED MATERIALS AND DEVICES FOR LASERS AND NONLINEAR OPTICS

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The field of molecular photonics is currently experiencing a significant revival of interest due to a number of reasons which will be outlined and exemplified in this talk:

1) Current and future developments of broadband telecommunication systems based on wavelength division multiplex (WDM) schemes are hampered by the current limitations of inorganic technologies. Initially proposed in the mid- to late eighties, electrooptic polymers have recently emerged at the forefront as strategic contenders with a demonstrated potential for high frequency electrooptic modulation well above 100 GHz unmatched by any other technology. The unlimited bandwidth of the non-resonant electronic hyperpolarizabilities at the root of the electrooptic effect and the favourable matching of optical refractive index with correspondingly low HF dielectric constants stand out as the main enabling features of polymers towards high speed operation. A number of passive and active functions such as filtering, switching or routing are now implemented in polymer devices benefiting from the increasing availability of low loss materials with high glass transition temperatures ensuring excellent stability and electrooptic coefficients of the same order as for Lithium Niobate or higher. Based on the adaptation to organics of otherwise well established processing steps (e.g. spin coating, microlithography, reactive ion etching), a full-fledged polymer based technology is now available to implement practically any conceivable optical circuitry on top of any mineral (e.g. Silicon and III-V semiconductors) or organic substrate, including flexible ones.

2) Whereas initial work in polymer based photonics was mainly focusing on channel waveguide architectures, more advanced structures are being increasingly explored such as microcavities based on planar, ring or disk geometries and photonic crystals made-up of the periodic repetition of micron-size optical features. Micro-laser sources with sizes in the range of 10 to 100 μm are now demonstrated and span the full visible to near IR range based on the outstanding luminescence efficiencies and good stability of novel organic dyes and conjugated polymers in the solid state.

3) The possibility to engineer advanced structures at the micron and sub-micron scales based on laser induced reorientation and displacement effects is likely to have a significant impact in this field all the way from fundamental light-molecule interaction studies to early device demonstrations. Indeed, quantum interference of different absorption pathways pertaining to coherent control techniques allow to engineer at room temperature (i.e. without resorting to temperature elevation as in the case of traditional poled polymers technology) unlimited patterns of optical properties for such applications as high density optical storage, nonlinear holography, photonic crystals, phase-matching schemes for optical wavelength conversion, parametric amplification or DFB laser structures. This domain is particularly illustrative of the virtues of soft matter photonics whereby photosensitive molecules can be reoriented at will in condensed phases so as to allow for the local imprint of desired optical properties.

4) Beyond the micron cale, fascinating and relatively untapped possibilities at the nanometer scale start to be accessible thanks to the increasing availability of advanced instrumentation techniques such as scanning tunneling, near field or confocal microscopy. The possibility to functionnalize polymer spheres of submicrometer size with luminescent semiconductor shells and to further arrange them in pre-organized 2-D and 3-D arrays and stacks with photonic crystal-like periodicity will be discussed in this perspective.

Real-time study of the dynamics of anisotropic phase separation in H-PDLCs.

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Abstract

We investigated the dynamics of formation of a reflection hologram in a photosensitive formulation containing pre-polymer and liquid crystal. Kogelnik's two beam coupling theory of an isotropic material shows a coupling coeffent, υ_p, of zero when p-polarized light is incident at an internal 45 degree to the diffraction grating. A broadband source was used as a probe to monitor the diffraction efficiencies (DE) during exposure for both s- and p-polarized light. The diffusion of the monomers sets up grating almost instantaneously during exposure as seen by the s-polarized light. The onset of a macroscopic ordering of the liquid crystal is observed when the DE of the p-polarized probe begins to grow. The real time experiments clearly establish the aniosotropic nature of the gratings formed. Experimental data shows evidence of shrinkage, beam coupling and the dependence of the onset of anisotropy with laser intensity.

Novel approaches to efficient generation, amplification and detection of THz waves

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Summary

THz waves have potential applications in bio-medical imaging, satellite communication, remote sensing, and non-destructive evaluation. However, so far emitters, amplifiers, and detectors in this domain are not yet operational at room temperature. We have investigated several novel schemes for THz devices. One can design and grow quantum-well and quantum-barrier dots to implement efficient THz emitters based on coherent oscillation and optical rectification. Quantum-well dots consist of an InGaAs/GaAs quantum well stressed by InP quantum dots. Therefore, there is an array of two-dimensional potentials within the quantum-well plane. If the quantum well is grown on the (111) substrate, a strained electric field can be induced in the quantum well. Therefore, center-of-inversion symmetry can be broken. These structures have certain advantages. Since quantum-well dots are inside the quantum well, surface and interface states can be avoided. One dimension can be precisely controlled. Due to the presence of built-in field in quantum-well dots, optical lithography and ohmic contacts can be avoided. It is easier to control coupling strength by using external electric field or growing coupled quantum-well dots. One can properly design these structures to reduce the scattering rate for the carriers by optical and acoustic phonons. These structures can also be modified to implement THz emitters and detectors based on resonant tunneling diode. The second mechanism for efficient generation of THz waves is based on transition radiation. One can diffusion-bond Ge/Si multilayers or grow epitaxial layers to form superlattices. If an electron beam transversely moves through these structures, electromagnetic waves can be generated from the interfaces based on transition radiation. If the thicknesses of these layers are properly chosen, a narrow-linewidth THz waves can be emitted due to constructive interference. Since the difference of dielectric constants can be very large, the emitted power can be quite large. One can use cyclotron resonance of electrons in vacuum and in semiconductors to detector millimeter waves or THz waves. These electrons can be excited by millimeter waves or THz waves. A laser beam can be scattered by these cyclotron electrons. By measuring the power of the Raman-scattered signal, one can detect these waves. We have considered forward and backward optical parametric oscillation and amplification, and difference-frequency generation for efficiently generating and amplifying terahertz waves in several second-order nonlinear optical materials. We have used a single crystal of CdSe as an example. We have also investigated GaSe, periodically-poled $LiNbO_3$ and $LiTaO_3$, and diffusion-bonded-stacked GaAs and GaP plates. The advantage of using birefringence in CdSe and GaSe is tunability of the output terahertz frequency. Furthermore, both CdSe and GaSe can be used to achieve the backward parametric oscillation without any cavity. On the other hand, in periodically-poled $LiNbO_3$ and $LiTaO_3$, one can take advantage of large diagonal elements of second-order nonlinear susceptibility tensor. In the diffusion-bonded-stacked GaAs and GaP plates, quasi-phase matching can be achieved by alternatively rotating the plates. The advantage of using coherent parametric processes is possibility of efficiently generating and amplifying temporally-coherent and narrow-linewidth terahertz waves. Compared with a noncollinear configuration, by using the parallel wave

35 words abstract:

$\chi^{(3)}$ enhancement of gold nanoparticles embedded in a dielectric host is determined at the surface plasmon resonance by z-scan measurements. Local electromagnetic field calculations give a better understanding of the experimental results.

Liquid Crystal based optoelectronic devices for communication networks

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The search for the best suitable materials for optical communication devices is an open problem and is one of the key point in the challenge for Terabit optical networks foreseen in the next 5-10 years. Organic materials like polymers, liquid crystals and liquid crystal composites may prove to be extremely useful in the realization of specific optical communication devices and in the design of systems for the optical networks.

We report here the preparation and characterization of novel isotropic polymer slab waveguides made by photochemical crosslinking, and their integration with a ferroelectric liquid crystal (FLC) in an integrated electro-optic modulator. The refractive index of the passive copolymer material in the device was controlled by the copolymer composition, and good agreement between measured and calculated effective indices for the guided modes of polymer films was obtained. The active FLC exhibited a refractive index change of $\Delta n \approx 0.10$ upon application of an ac voltage of ± 30 V, giving an electro-optic modulation of the input TE0 -mode with a contrast ratio of 11:1 and only a small polarization conversion into TM output light. The time response of the device was in the typical range of FLC's.

Keeping these results as a starting point, we explored the possibility of using FLC for making active devices based on Bragg gratings in planar waveguides. Two different devices have been envisaged: an integrated electro-optic switch and a continuously tunable filter.

The first device exploits the fast and bistable switching of *FLC's* in the Surface Stabilized structure. The principal advantage of this device is its spectral signature, which allows overcoming the problems of intensity dependent devices.

The soft-mode of *smectic A** FLC's, allowing a continuous modulation of the extraordinary refractive index, has been utilized in order to design an integrated tunable optical filter in the wavelength range of interest for optical communications. The principal advantages of such device include fast tuning speed, wide tuning range, low power dissipation, and low cost.

Excited state absorption and reorientation in polymethine and squarylium dyes.

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Abstract

Over the past decade, the strong excited-state absorption exhibited by some organic molecules has attracted considerable interest for possible device applications. However, not much is known about the nature of this excited state absorption. Here we describe a detailed theoretical and experimental study of the excited states of polymethine and squarylium dyes. Picosecond, polarization-resolved excite-probe experiments reveal that orientational relaxation provides an important decay channel, surprisingly even in solid state hosts. Subpicosecond transient absorption and anisotropy excitation measurements were performed in several polymethine and squarylium dyes in ethanol solutions and polymeric host over the spectral range 400 - 1500 nm. A variety of nonlinear effects including saturable absorption, reverse saturable absorption and gain were observed and analyzed. We observe strong excited-state absorption (ESA) in all dyes in the range 450 nm - 600 nm. We also report the first observation of additional ESA bands in the near IR range. To determine the nature of these transitions, measurements, we performed quantum-chemical calculations. The relationship between ESA spectra of organic dyes and their molecular structure are discussed.

Multi-photon microscopy for micro/nano photofabrication, high-density data-storage, and cell imaging/surgery

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Abstract:
Multi-photon microscopy has been applied to three-dimensional micro/nano fabrications with photosensitive materials. The principle and the experimental setup will be shown with many of experimental results including three-dimensional complex photopolymerzed structures, rewritable multi-layer photoisomer data-storage, and multiphoton-stimulated Calcium-ion imaging in cells.

Summary (200 words):
The combination of the latest femtosecond laser technology with the advanced laser scanning microscope technology makes it possible to fabricate subwavelength structures in various photosensitive materials in three dimensions. Nonlinear spectroscopy or multi-photon process is the underlying physics. Three-dimensional complex micromachines are fabricated at subwavelength precision with two-photon photopolymerzation [1,2], that can not be made with a conventional photolithgraphic technology. Fifty of layers of digital data are recorded in a 150 μm-thick photoisomer-doped PMMA by two-photon photoisomerization, and are read without crosstalk between layers with confocal microscope optics [3-5]. Multi-photon process has been also applied to the genaration of damage in the cells, collagen, and other biological tissues [6], as well as to the stimulation of Calium ion wave generation in cells [7]. Subwavelength imaging of molecular vibration in a cell was also successfully made by mixing two beams of diffferent wavelengths of femtosecond (or picosecond) near infrared laser to generate coherent anti-Stokes Raman scattering [8]. The talk will extend to fabrication of selfgrowth of fiber structures and intereremetric production of photonics crystals [9,10].

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Numerical Analysis for Characteristics of QW-HPT/QW-LD Optoelectronic Integrated Device

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Recently, there has been increased interest in optoelectronic integrated device (OEID) which constitute the cornerstone in the various optical signal processing systems and optical computation. OEID can also be used for parallel signal processing, wavelength conversion, light amplification, and optical switching. A vertical and direct integration of a light-detecting and a light-emitting device, such as heterojunction phototransistor (HPT) and a laser diode (LD) is an effective method to obtain optoelectronic integrated device. The input light is converted to photo-generated carriers through the light-detecting device, which drives the LD to on state. Integration of HPT over LD gives rise to new phenomena such as internal electrical and optical feedback between LD and HPT, which have been utilized to get new functions such as optical amplifier, optical switch, and optical bistable.

To increase the response speed of device, we propose an OEID monolithically integrated by a quantum well structure HPT with n-p-i-n configuration over a quantum well laser diode (QW-LD) for the first time. The schematic structure of device is shown in Fig. 1. The grown process can be carried out by metal organic vapor phase epitaxy (MOVPE) or a combination of liquid-phase epitaxy (LPE) and MOVPE. The incident light is absorbed in base and multiple quantum well-i region between base and collector and convert to photo generated current.

A physical model for operation of this quantum well optoelectronic integrated device (QW-OEID) is develpoed to investigate quantitatively dynamic and static response of device and its out put characteristics. Base on the model, considering the effect of positive internal optical feedback, optical gain and rise time of the QW-OEID is computed numerically The model derives two different modes of operations for device: amplification mode for small optical feedback, and switching mode for high feedback.. The effect of several parameter such as HPT optical conversion gain, HPT cut-off frequency, HPT base width, QW-LD gain, and optical feedback coefficient on operation of device are investigated quantitatively.

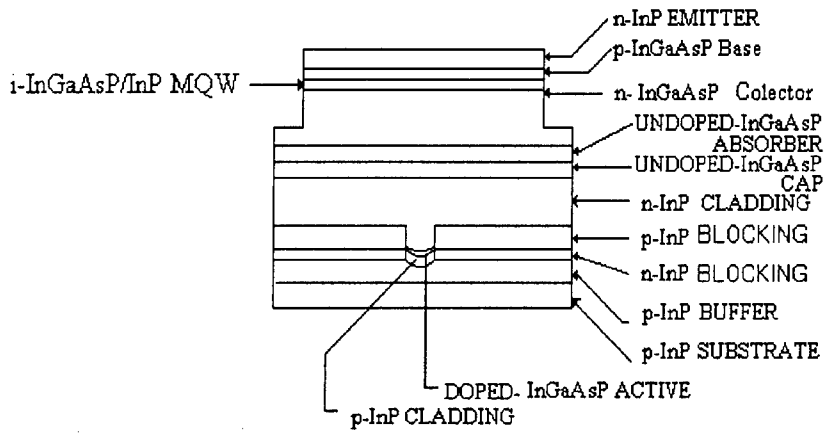

Fig. 1: Schematic structure of optoelectronic integrated device

Bistable nematic devices: surface and bulk melting

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The behavior of orientationally ordered fluids, restricted by symmetry breaking surfaces, significantly deviates from bulk systems. Usually, the anchoring effect is described as a small and completely reversible surface director deviation under applied external constraints. This "elastic" approximation works well for the well known and largely employed strong anchoring cases. In the weak anchoring case, the experiments show more complicated behaviours. Surface melting induced by external fields is another challenge for the liquid crystal research.

All presently used liquid crystal displays are based on texture change in thin cells under application of a suitable external field. These textural changes appear above a low voltage threshold characteristic of textural bulk "bifurcation".

Three novel electro-optic devices, which are far beyond the present liquid crystal technology, are, at present, in the developing stage in Europe. They are based on surface anchoring breaking, i.e. on surface bifurcation. They deal with topological changes of the liquid crystal texture that are not compatible with fixed boundary conditions. They use nematic materials and show an intrinsic texture bistability.

In particular conditions, the topology change of the nematic texture can be achieved also by a "bulk" melting induced by an external field. Hence, it could exist a competition between surface anchoring breaking and bulk melting to obtain a controlled textural switch in bistable nematic cells.

A delicate analysis of experimental results shows that usual theoretical descriptions are unable to justify the observed orientational transitions.

INFRARED THIRD ORDER NONLINEAR OPTICAL CHARACTERIZATION OF POLYPHENYLENE VINYLENE DERIVATIVES

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Polymers have been the subject of intense research during the past decade, as third-order non linear optical materials for photonic applications [1]. However, most optical studies have been performed so far in the visible spectral range, while optical communications are focused on the infrared range around $\lambda = 1.5$ μm. In particular, the conjugated polymer poly(1,4-phenylene vinylene) (PPV) and its soluble derivatives were so far studied as visible light emitting materials and for photovoltaic applications.

In this work we report the results of the experimental characterization of the third-order nonlinear response of PPV thin films. The χ^3 coefficient has been evaluated by Third Harmonic Generation (THG) using Maker fringes technique [2], thus measuring the fast electronic contribution to the nonlinear coefficient. A tunable Q-switched laser source have been employed, allowing to perform measurements for fundamental wavelengths spanning the infrared range from $\lambda = 1.2$ μm up to $\lambda = 2.1$ μm. In this way, the dispersion of the THG coefficient both in and out of resonance has been analyzed and compared to absorption spectra.

The experimental setup has been engineered in order to overcome laser source instabilities and to avoid any spurious contribution due to air. Characterizations have been performed on Poly[1,4-phenylene-1-phenyl vinylene] **P-PPV**, poly[1,4-phenylene-1,2-di(4'-phenoxyphenyl) vinylene] **DPOP-PPV**, poly[2- methoxy -5- (2'-ethylhexyloxy)-1,4-phenylene vinylene] **MEH-PPV** and poly[2,5-dioctyloxy-1,4-phenylene vinylene] **DO-PPV**. The 0.1 – 0.2 μm-thick films were prepared by spin-coating of 0.8 % polymer solutions in o-xylene onto BK-7 glass substrates under rotation rate at 3000 r.p.m.

The measured χ^3 values strongly depend on the effect of the substituents of the PPV derivatives, which probably influence the planarity and π-conjugation in these polymers. The highest value $\chi^3 = 8 \times 10^{-11}$ esu has been observed for MEH-PPV of high molecular weight (Mw 230.000), at $\lambda = 1.77$ μm. A spectral shift between the χ^3 and absorption peaks has been noticed in all samples, being attributed to a disorder in the distribution of the π-conjugation length, taking into account that long rigid π-conjugated sequences are particularly effective for large χ^3 close to three photon resonance. No change in the nonlinear optical properties of samples was registered after heating at 70°C in air for 10 days and exposure to intense infrared light. Infrared absorption coefficients as low as $\alpha = 0.2$ cm^{-1} have been measured in solution, therefore increasing the interest in the properties of this promising class of polymers, in view of possible applications in photonic devices for telecommunication systems, also taking into consideration their good processability.

The authors wish to acknowledge Prof. O. Efimov, Dr. I. Yakushenko and Dr. V. Belov from Institute of Chemical Physics (Russian Academy of Sciences) for supplying with PPV derivatives and helpful discussion.

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Photoinduced phase transition in a liquid crystal – azobenzene derivative system.

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Reversible photochemical reactions have been known to influence the stability of liquid crystalline phases: for example, nematic-to-isotropic phase transitions can be triggered by *trans*-to-*cis* photoisomerisation of azobenzene derivatives (e.g., [1-4]). However, no examples of a reverse process - photoinduced formation of a nematic phase have been reported in the literature.

The main aim of the work was to study a photoinduced isotropic-to-nematic phase transition in pentylocyanobiphenyl (5CB) doped with 4-fluoro-4'-methoxyazobenzene. The samples, containing a few percent of the dye, were thermally stabilised and observed with an Olympus polarising microscope at constant temperatures, controlled to within 0.1°C. The nematic-to-isotropic transition could be observed upon irradiation with 366 nm light at temperatures substantially (over 10°C) lower the thermodynamic phase transition temperature, the rate of formation of the isotropic phase depending on the light intensity. The reverse reaction was found to be driven by the 440nm light, its rate depending on the intensity of the illumination. Separate experiments ruled out a possibility of the isotropic-to-nematic transition being thermally driven.

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Nonlinear Optical Responses of Antifferromagnetic Insulators

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The parent crystals of high temperature superconductors are antiferromagnetic (AF) insulators below room temperature, which comes from strong correlation of (3d) electrons in Cu and charge-transfer between Cu (3d)- and O (2p)- orbitals. In order to describe the electronic structure and nonlinear optical responses of these crystals with layered Perovskite structure, we have developed excitonic cluster model so as to take into account the strong correlation effect and large charge transfer on the same footing [1,2,3]. First we could resolve a mystery about resonance-enhancement of two-magnon Raman scattering in terms of the present model [2]. This can explain the absence of resonance-enhancement of this two-magnon Raman scattering due to the destructive interference of two lowest excitons composing the strongest absorption peak on the lowest energy in La_2CuO_4 and YBa_2CuO_6. Second we observed strong third-harmonic generation (THG) in visible region from La_2CuO_4 [4], and could understand the fine structures as quantum interference among two-photon resonant THG to even modes of the exciton [3] and three-photon resonant THG to the odd modes [2].

Ferroelectric (FE)-AF crystals $RMnO_3$ (R=Y, Ho, Er) with a hexagonal structure show interesting two-photon resonant second-harmonic generation (SHG). Two lowest excitons in the visible region are found to contribute to SHG constructively in $YMnO_3$ and destructively in $ErMnO_3$ depending upon the AF spin-configuration [5]. We found also the clamping of two order parameters FE and AF induced at the domain boundary [5].

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NEMATIC LIQUID CRYSTALS A NICE ENVIRONMENT FOR SPATIAL SOLITONS AND THEIR APPLICATIONS

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Spatial solitons, i. e., guided field eigen-distributions of nonlinearly self-induced waveguides, can be formed in nematic liquid crystals by a reorientational response, overcoming diffraction through a refractive index increase.[1-3] Contrary to ideal Kerr media, such increase is saturable and nonlocal, [4-5] and can prevent filamentation and catastrophic collapse even in the fully three-dimensional case.[6] To sustain a soliton, a linearly polarized optical field exerts a torque on the (pre-aligned) induced molecular dipoles of a positive uniaxial cystal, orienting them towards the optic axis. This can be obtained at low powers by using dye-dopants to overcome the Fréedericks threshold,[1] or launching a combination of TE and TM polarizations in planar waveguides.[2] To reduce thermal effects, another approach consists in lowering the required excitation by introducing a pre-tilt to the molecules in a planarly-anchored nematics, as to form an angle smaller than _/2 between them and the linearly polarized E-field in the beam. [3]

Based on the latter technique, using an external low-frequency voltage to control the director pre-orientation, we discuss on 3-D spatial solitons formation and propagation in planarly-anchored undoped nematic crystals by mW laser excitation. Since the liquid crystal has a non-instantaneous and non-local response, it can average out rapid intensity variations in the beam. This feature allows to employ beams with a low degree of spatio-temporal coherence. To assess this property, we introduced a rotating random diffuser in the path of a multimode Argon laser at 514nm. When properly polarized and propagating in a biased cell, at excitations slightly higher than the coherent counterpart, the beam formed an "incoherent" spatial soliton. The optically-induced waveguide could confine not only a weaker (<0.1mW) coherent (copolarized and collinear) signal at another wavelength (633nm), but also one traversing the same diffuser employed for the pump.

To explore the potentials of such an all-optical waveguide, we launched a weak "signal" beam at an angle to the intense (pump) excitation. At longer wavelengths a co-polarized signal was "trapped" in the soliton for tilts as large as ± 3°, with no trapping in the other polarization. Finally, we

exploited the wide angular acceptance of the soliton to obtain all-optical steering and readdressing. Varying the relative angle between pump and signal beams at the input, we verified that the signal followed the path of the self-confined pump and was effectively readdressed at the output plane. For propagations of 1mm, the angular steering led to output shifts as large as 30_m, equivalent to several resolvable spots or spatially routed channels.

These preliminary results demonstrate wavelength independent phenomena observable with LED sources, and pave the way to novel generations of optically controlled spatial demultiplexers and readdressable interconnects for transparent optical networks.

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Nonlinear organic photorefractive polymers and their applications

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Photorefractive polymers are among the most sensitive nonlinear optical recording materials. They exhibit large refractive index changes when exposed to low power laser beams. Discovered and studied for several decades mainly in inorganic crystals and semiconductors, the photorefractive effect has been discovered in organic materials in the 1990s. In recent years, guest-host polymers with nearly 100% diffraction efficiency, increased shelf-life, and ms response-times could be demonstrated.

Here, we will report on the fabrication of photorefractive polymer composites using injection molding (see Fig. 1). This work is a proof-of-principle demonstration that photorefractive polymers can be fabricated using mass production techniques. For our experiments we formulated a photorefractive composite that was known to have good phase stability properties and a high dynamic range. The inert polymer was a commercial birefringence-free acrylic resin, doped with the photorefractive chromophore 2, N, N-dihexylamino-7-dicyano-methyl-idenyl-3, 4, 5, 6, 10-pentahydro-naphthalene (DHADC-MPN), that provides simultaneously transport properties and electro-activity. The composite was plasticized using diphenyl isophthalate (DIP), and sensitized using (2,4,7-trinitro-9-fluorenylidene)-malononitrile (TNFDM). The field dependence of the diffraction efficiency measured in steady-state four-wave mixing experiments performed at 633 nm is shown in Fig. 2.

We will also report on new photorefractive polymers that are sensitized by nonlinear absorption. Regular photorefractive polymers are doped with a sensitizer that often forms a charge transfer complex with the transport moieties. This complex provides optical absorption at the operating wavelength. In contrast, nonlinear photorefractive materials are transparent for low power cw beams and become absorbing at high intensity through multi-photon absorption. In our case, two-photon absorption was provided by the electro-active chromophore. New properties associated with nonlinear absorption including non-destructive read-out and photon gating will be discussed.

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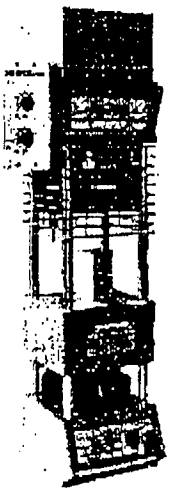

Fig. 1: Photograph of the Morgan Industries G55-T 22 ton vertical travel press used to fabricate by injection molding samples of photorefractive polymers with various shapes and thickness.

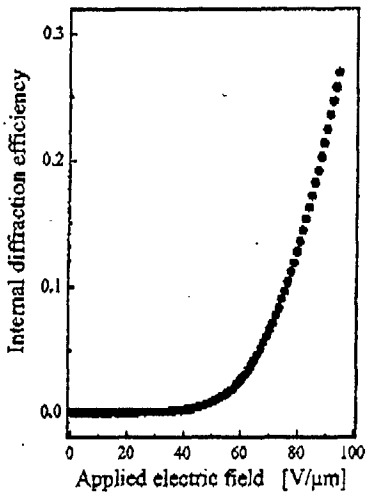

Fig. 2: The diffraction efficiency of an injection-molded photorefractive polymer composite sample as a function of applied field.

Recent advances in photorefractive solitons

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After a brief hystorical introduction, the state-of-the-art of photorefractive solitons is reviewed. This research topic, far from having reached saturation, is still expanding along different directions, that is theoretical, experimental and applicative. In particular, the availabilty of new photorefractive crystals exhibiting a room-temperature phase transitions between the asymmetric ferro-electric phase and the symmetric paraelectric one has resulted in the observation of completely new class of phenomena.. On the theoretical side, the still open problem of how to generate circularly symmetric propagation by exploting a basically anisotropic effect is eventually exhibiting some real progress. Also, the actual experimental investigation of collisions and inhomogeneous forces between solitons of different dimensionality (a self-trapped stripe and a round soliton) has stimulated the study of new analytical techniques for finding solutions of nonlinear differential equations describing the simultaneous formation and interaction of one-dimensional and two- dimensional localized coherent structures. New soliton-formation stabilization processes, connected to the beam behavior in a nonstationary external-bias field, have also been recently described. The combination of charge separation induced by the formation of a single photorefractive screening soliton and an applied external bias field in a paraelectric has been shown to lead to a family of useful electro-optic guiding patterns and properties. The observed phenomena represent an important step in the achievement of feasible soliton-based components whose switching dynamics are limited only by capacity-charging times, as in all other electro-optic devices.

Photochromic Liquid Crystals; From In-Plane Switching to Optical Recording

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Certain organic materials, called photochromics, undergo *trans-* to *cis-* isomerisation under light illumination (photoisomerisation) [1]. The changes in the molecular structure of these materials that take place during the photoisomerisation process might result in significant changes of their physical properties. The photoisomerisation process, if there is no chemical reaction or material degradation under the light illumination, might be reversible. Some photochromic materials that also exhibit liquid crystalline phases have recently become very attractive materials for applications since their bulk and surface properties can be efficiently controlled by light.

This talk will be focussed on changes in bulk and surface properties of some photochromic liquid crystals that take place under light illumination [2]. Here will be presented several light-induced effects that seem to be promising candidates for implementation in different kinds of liquid crystal devices for photonics. Such examples as optical recording, optically controlled light modulators and switches with photochromic liquid crystals will be given.

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Dynamics of liquid crystal azimuthal anchoring at polyvinylcinnamate interface measured *in-situ* during polarized UV light irradiation.

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The aim of the present paper is to study dynamics of LC azimuthal anchoring at an LC - photosensitive polymer interface during the irradiation of the latter made *in-situ*, that is directly in a cell filled with a liquid crystal. Such a process has some important peculiarities which shed light on the mechanism of the anchoring.

Before irradiation, the planar orientation throughout the bulk of the cell was induced by rubbing a polyimide covered rear ITO interface. Then the front ITO surface covered by photopolymer (polyvinylcinnamate, PVMC) was exposed to the linearly polarized UV light with the electric vector at various angles with respect to the initial director orientation. Due to reorientation of the easy axis at the photopolymer interface a twisted nematic (E7) structure was created. An automated *in-situ* measurements of the irradiation induced twist angle have been carried out.

The evolution of the director reorientation has been described in terms of a competitive action of two surface anchoring: the initial one due to adsorption memory of the LC bulk alignment irrelevant of irradiation and the other one induced at the PVMC interface by polarized UV-light illumination The photoinduced azimuthal anchoring has been shown to reach values of ~20-23 merg/cm^2. The value of the adsorption part of azimuthal anchoring is about~7 merg/cm^2, that means that a weaker azimuthal anchoring is hardly reachable in this system. The time of the director reorientation is controlled by a slow adsorption-desorption process and is of the order of an hour while the birefringence in the PVMC-film under study is induced within few seconds.

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Permanent polarization gratings in photosensitive Langmuir-Blodgett films

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Abstract

An investigation was carried out on thin permanent phase polarization gratings recorded in Langmuir – Blodgett films using two interfering orthogonal circularly polarized Ar-ion laser beams. The films are composed of amphiphilic azo-dye molecules and manifest an extremely large value of the photo-induced optical anisotropy, $\Delta n \approx 0.36$. They give long time stability (more than one year) and high photoinduced birefringence that provides high diffraction efficiency of the grating The experimental results are in excellent accordance with the theoretical model based on the Jones matrix representation of the polarization pattern [1].

We show how the polarization gratings permanently stored in these films are very interesting devices for polarimetric applications, in particular as components of a spectrophotopolarimeter.

G. Cipparrone, A. Mazzulla, S.P. Palto, S.G. Yudin and L.M. Blinov, Appl. Phys. Lett. **77,** 2107 (2000)

Optically addressed spatial light modulators and their applications

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Abstract:
Optically addressed spatial light modulators (OASLMs) are now important devices for system developers. This presentation covers two contemporary display applications, and latest results on the devices.

Summary:
Optically addressed spatial light modulators (OASLMs) hold great promise for high resolution and 3D display systems [1,2]. They have a multilayer structure which is readily fabricated in a well equipped research laboratory. The amorphous silicon/ ferroelectric liquid crystal spatial light modulator has the following layer sequence: ITO; alignment layer; ferroelectric liquid crystal; alignment layer; amorphous silicon; ITO (Fig. 1). Since it was first demonstrated [3], there has been very little work on tailoring the device for specific applications. High resolution, large format displays require high resolution, bistable, multiplexable devices with high optical efficiency. The resolution of the first devices was 70 lp/mm at 10% MTF, when the drive conditions were optimised for maximum contrast [2]. This deteriorates when a dielectric multilayer coating is introduced between the amorphous silicon and FLC, in order to improve the optical efficiency of a reflective device [4]. Bistable liquid crystal response was gained by the use of SCE13 liquid crystal with a silicon oxide alignment, which was electrically treated to produce the quasi-bookstack geometry [5].

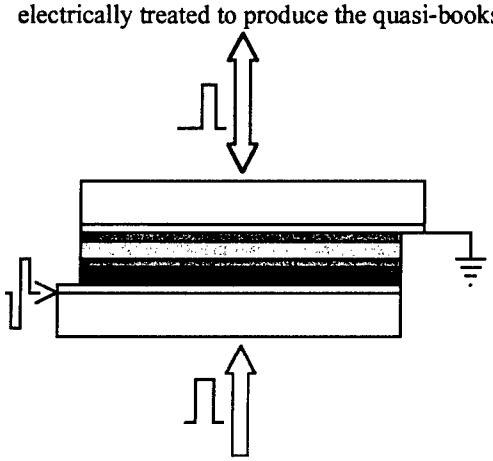

Fig. 1 Layered structure of an OASLM.

In our laboratory, we are systematically establishing the optimum process parameters, using the construction of the first FLC OASLMs as a guideline. We have constructed an optically addressed spatial light modulator using a 2 micron layer of amorphous silicon as the photoconductor, and a 1.5 micron layer of an experimental liquid crystal mixture based on siloxane oligomers. A rubbed nylon alignment produces cells with large switching angle (typically 57 deg) and bistable behaviour is established by use of electrical post-processing [6]. The device is addressed both electrically and optically. A bipolar electrical waveform addresses the device (Fig. 1), the first pulse RESETS the modulator to a uniform optical response and the second pulse SETS the modulator to the spatial information on the write beam. The write beam addresses the photoconductor side of the device and the read light, which is reflected from the liquid crystal side of the device, is synchronized with the SET cycle of the electrical waveform. Bistability is currently adequate for the envisaged applications, and we are proceeding to test the resolution of these devices.

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LOCALIZED STRUCTURES IN A NONLINEAR OPTICAL INTERFEROMETER

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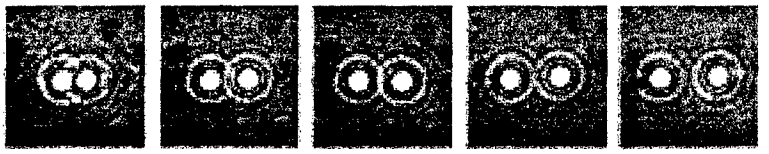
We report a study about the localization of spatial dissipative structures in a nonlinear optical interferometer.

After briefly reviewing the conditions for localization of the patterns rather than the appearence of structures extended across the whole wavefront, we investigate dependence of the observed structure stability and shape on experimental parameters.

We show how the interactions between these "dissipative solitons" lead to the formation of several molecule-like bound states, formed by pairs of localized structures.

The dependence of the main characteristics of these bound states on the system parameters is measured and discussed.



Example of bound states formed by pairs of localized structures

Self-assembled vs. interface quantum dots: the effect of thermal coupling on optical properties

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In spite of the great interest on zero-dimensional semiconductor structures stimulated by their potential for new photonic and electronic applications, a consensus on a general modelling of their optical properties has not been reached yet. Here we report experimental results on the temperature and power density dependence of the photoluminescence (PL) for two different kinds of quantum dot (QD) systems, namely: (a) a set of InAs/GaAs self-assembled QD (SaQD) samples and (b) a set of $In_xGa_{1-x}As/In_yGa_{1-y}As/GaAs$ asymmetric quantum wells (AQW) showing spontaneous QD formation at the In-alloy/In-alloy interface, i.e., natural quantum dots (NQD). The SaQD samples consist of both single and stacked layers with an InAs coverage ranging between 2.7 and 3 ML. The AQW samples have been obtained with an intentionally abrupt change of indium composition inside the well ($x = 0.149$, $y = 0.064$) [1].

Both QD systems show a composite peak in the low-temperature PL spectra and an incipient saturation by increasing the excitation power density. Moreover, PL spectra exhibit an unusual temperature dependence peculiar for each type of QD. The SaQD PL full-width-at-half-maximum (FWHM) displays an initial reduction down to a minimum at around 100 K, followed by an increase at higher temperatures, while the NQDs manifest an almost constant FWHM over the whole temperature range. In addition, the SaQDs display a PL red-shift larger than that of InAs bandgap. On the contrary, we observe a reduction of the Stokes shift with temperature in the AQWs samples (i.e., a relative PL blue-shift).

As reported by several authors, the PL peak position of a single QD, whether a NQD or a SaQD, shifts with temperature as the bulk bandgap does. Therefore, an explanation of the PL spectra evolution, observed probing a large QDs population, should arise from some collective mechanism. Indeed, the measured SaQD PL behavior agrees well with existing models [2]. We will discuss how our experimental findings can be treated within a similar framework. We suggest a steady-state model that takes into account both carrier thermalization and the role played by different escape and retrapping channels. The SaQD PL data may be satisfactorily reproduced by a scheme in which uncoupled QD states share a common channel for carrier exchange (wetting layer). However, to account for the NQD PL data, it is necessary to add to the former scheme a direct coupling mechanism between QD states. We will discuss the role of the AQW free exciton as the main coupling channel.

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Spatial Modulational Instability in Planar Waveguides

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Abstract
When intense optical beams propagate through media characterized by self-focusing or beam narrowing optical nonlinearities, modulational instability (MI) can occur. This results in the generation of periodic structures whose periodicity depends on the nature of the self-focusing mechanism, the intensity of the beam and the Fourier components present at the input. If generated from noise, the Fourier component with the highest gain (for the input intensity) is amplified the most and dominates the output beam periodicity. If the input beam is seeded with a periodic modulation, then that period is amplified.

We have investigated both cases in Ti:in-diffused $LiNbO_3$ waveguides near their condition for birefringent second harmonic phase matching. The beam narrowing inherent to multi-wave mixing led to the generation of periodic patterns whose periodicity decreased with increasing power, in excellent agreement with theory. In the second set of experiments, the input beam was spatially modulated by producing an interference pattern with an intersecting weak probe beam. This allowed the MI gain coefficient to be measured as a function of periodicity, input intensity and phase-mismatch for the first time.

Experiments in Kerr AlGaAs waveguides may also be discussed if the data is available.

Next Rays? T-Ray!

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During the last twenty years, innovations in microwave electronics (such as internet and wireless communication) and in photonics (such as semiconductor lasers and fiber optic communication) have changed the way we function and communicate in and with the world around us.

Terahertz (THz) radiation, which is electromagnetic radiation in a frequency interval from 0.1 to 10 THz, is the next frontier in physics and technology. This band occupies a large portion of the electromagnetic spectrum between the infrared and microwave bands. However, compared to the relatively well-developed science and technology in the microwave and optical frequencies, basic research, new initiative and advanced technology developments in the THz band are very limited and remain unexplored.

THz science will impact information technology and biotechnology. Specifically, it will have enormous potential applications in communications, imaging, medical diagnosis, health monitoring, environmental control and chemical and biological identification. We expect that the research in the THz band in the 21st century will become one of the most promising research areas for transformational advances in physics, as well as in other interdisciplinary fields. I will highlight the recent advances of novel optical materials to impact terahertz wave sensing and imaging applications.

INNOVATIVE PHOTOREFRACTIVE MATERIALS

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We present here two different classes of new photorefractive materials. In the first one a single low molecular mass molecule exhibits all the functions necessary for photorefractive behaviour. The active species is an highly efficient cyclopalladated molecule which contains a palladium(II) center bonded in a square planar geometry. Most of the results shown have been obtained using a compound which results from the cyclometalation of azobenzene and from the coordination through oxygen and nitrogen atoms to the 2-hydroxy-4-(*n*-hexyloxybenzylidene)-4'-*n*-hexylaniline. We performed two-beam coupling experiments by overlapping on our samples two He-Ne laser 'writing' beams which were either s or p polarized. For p-polarized beams we observed a monotonic increase of the gain up to a value of $\Gamma = 230$ cm^{-1} at E = 28 V/μm, which is among the largest measured to date at such low fields in organic materials. The gain then saturates and it is constant for higher fields. For s-polarized beams the gain is much smaller and of opposite sign. This indicates a substantial orientational contribution to the refractive index grating. To estimate the amplitude of the index grating we also performed degenerate four-wave mixing (DFWM) experiments. The diffraction efficiency shows a maximum for an applied field E ~ 11 V/μm and it decreases for higher fields, becoming a constant at E ~ 27 V/μm, the same field for which the gain saturates. From DFWM experiments it is possible to extract the amplitude of the refarctive index modulation Δn. This is done by using Kogelnik's coupled-waves model for slanted dielectric thick transmission gratings. From the model we obtain $\Delta n = 4.7\ 10^{-3}$ at 11 V/μm, where we observe the maximum efficiency. For fields higher than 27 V/μm we reach a constant value of $\Delta n = 9.3\ 10^{-3}$. Measurements of photocurrents have also been performed in materials of the same class. The photocurrent depends linearly on the square of the applied field and it reaches a value of $5\ 10^{-13}$ cm Ω^{-1} W^{-1} at 95 V/μm.

In the other class materials liquid crystals are used. The first photorefractive mesophases were bulk nematics or dispersions of nematic domains in photoconducting polymers but the variety of phases which exhibit liquid crystalline behaviour offers many other possibilities which are worth of investigation. Among them we have the chiral smectics. While the response of a nematic to an electric field is quadratic in the field and is due to the anisotropy of its dielectric constant, chiral smetcics can also have a linear response due to the interaction of the field with a spontaneous (ferroelectric) or induced polarization. In both cases the net result is a reorientation of the phase optic axis but the underlying physical processes are very different. The drive behind the development of such smectic photorefractive materials lays in their fast reorientation times, which can be in the range of 10 μs instead of the 1-10 ms typical of most nematic devices. In order to optimize the photorefractive performance of smectic materials it is necessary to understand how the index modulation depends on a variety of parameters. We present here a simple model to calculate the refractive index modulation associated with the director reorientation in photorefractive S_A^* and S_C^* phases. The model takes into account several material parameters, as well as the direction and the polarization of the incident beams. We first obtain the local director orientation under the simultaneous influence of an applied and a photogenerated electric field and we then complete the task of obtaining the refractive index modulation taking also into account the polarization and direction of the incident light and the orientation of smectic layers. A comparison with experimental data will be illustrated.

Title: Polymer Cholesteric Liquid Crystal Flakes for Display and Other Electro-Optic Applications

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Abstract:
Polymer cholesteric liquid crystal (PCLC) flakes retain optical properties like selective reflection and circular polarization of light associated with low molecular weight cholesteric liquid crystals. Unlike typical mesogens, however, PCLC flakes, which resemble particles of the order of tens of microns in length, are robust and stable over a wide range of temperatures. When suspended in a host fluid, PCLC flakes can be manipulated in the presence of an electric field to control their motion. As a PCLC flake rotates, its selective reflection color is both shifted and diminished. Thus it is possible to "switch" a flake on and off. To date we have recorded flakes rotating from an "on" to "off" state within 80 ms in the presence of a 3 V/μm electric field. The response time and applied field are comparable to those of other large particle display technologies. The shape of the flake has proven to be crucial in obtaining consistent and reproducible motion, and new methods for developing uniformly shaped flakes are being investigated. Using flakes with selective reflection bandwidths throughout the visible spectrum, it could be possible to create color displays. Thus PCLC flakes open the possibility for switchable, yet stable, optical devices with properties unique to liquid crystals.

Acknowledgment:
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Azobenzene-derivatized Polyimides as Photoreactive and Alignment-Layers

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Various azobenzene-derivatized polyimides have been synthesized and used for the build-up of photoreactive multilayer assemblies. To this end, we firstly evaluated the monolayer properties of these ampliphilic side-group polymers at the water/ air-interface. After transfer to a solid substrate the resulting Langmuir-Blodgett-Kuhn multilayer assemblies were characterized by surface plasmon- and waveguide spectroscopy, by X-ray reflectivity, and by UV-vis spectroscopy. Functional properties that were assessed were, in particular, the photo-reactivity associated with the photo-isomerization of the azobenzene chromophores. Possible applications are for optical switching in integrated optics devices and tunable alignment layers in the construction of liquid-crystal cells.

Novel Nonlinear and Photonic Crystal Strucures with Liquid Crystals

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ABSTRACT

Refractive index change is the fundamental parameter responsible for the working principles of almost all types of optical devices, including for examples optical logic gate, router, switches, holograhic gratings. In most current devices, the required index change is generated in some electro-optical materials through the Pockell cell effect. It is also possible to optically generate the required refractive index modification in some glasses using UV radiation. However, the resulting index change is permanent, and applicable for a fixed wavelength. For reconfigurable and broadband application, transient refractive index changes produced by optical Kerr effect or other nonlinear optical processes are more desitrable. .

Nematic liquid crystals (NLC) possess very broadband birefringence and transparency [from the 400 nm to 20 microns spectral region]. Because of this and other unique physical properties, compatibility with integrated optics and nanoelectronics, and acceptable response speeds, NLC's are widely used in various optoelectronics and nonlinear optics applications [1,2]. We have recently conducted a quantitative study of the optical nonlinearities of nematic liquid crystals in the near IR communication spectral region. It is found that in this region, the observed nonlinearities rank among the largest of all known materials, and allow us to observe several switching and modulation processes with low power lasers.

In the quest to develop next generation photonic switching devices, recently there have been much interest in developing photonic bandgap cyrstals, which comprise 1-D, 2-D or 3-D array of materials of different indices of refraction. In particular, 2-D and 3-D structures with nanometer pore infiltrated with nonlinear optical materials are highly promising since the resulting photonic bandgap crystals are tunable [2]. We report here recently obtained nonlinear diffraction and director axis alignment results from nematic crystals confined between cell windows with sub-wavelength surface gratings, and 2- and 3- D photonic crystals or nano-particles network impregnated with supra-nonlinear nematic liquid crystals.

ACKNOWLEDGEMENT
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NOMA 2001, Cetraro, May 20th-26th

Local field effects in resonators: from optical limiting to generalized bistability in single-mode laser dynamics

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Optical bistability can occur in two-level atoms without any external feedback. If the atomic density is so high that dipole-dipole interactions are exalted, Local Field Correction (LFC) is necessary. For relatively high dipole-dipole interactions, the material can then present a high- or a low-excited state when illuminated by a resonant incident wave : it is one example of the so-called Intrinsic Optical Bistability (IOB).

We first investigate theoretically the behavior of a dense medium inserted in a passive cavity. We show that LFC can lead to optical limiting. This property is interpreted as a perfect compensation of the input intensity increase by additional losses resulting from both nonlinear absorption and dispersion. This process occurs for incident intensities leading to the presence of a spatially localized first order phase transition between the two phases of high and low excitation within the sample.

We further investigate the influence of relatively low dipole-dipole interactions in homogeneously broadened single-mode laser dynamics. We first demonstrate that the second laser threshold (Lorenz-Haken instability threshold) can be significantly reduced when the Local Field Correction is taken into account. The routes to chaos are then identified for different values of the detuning and of the LFC. We show that LFC reduces the range of instability and leads to a restabilization of the off-state for very large negative frequency detunings. In addition generalyzed bistability can then be observed between the unstable chaotic regime and the off-state.

Nonlinear Optics for the Generation and Detection of Ultrafast Electric Fields: Techniques and Applications

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Abstract

Femtosecond lasers have had a dramatic impact on many areas of nonlinear optics. In this presentation, we will discuss the use of nonlinear optics to produce controlled electric field waveforms on the femtosecond time scale and to measure their temporal evolution. The former may be accomplished by optical rectification; the latter may be achieved by means of the second-order nonlinear response (electro-optic measurements) or through the third-order response (field-induced second harmonic generation). The combination of these approaches provides a powerful means for time-domain far infrared spectroscopy.

In this presentation, we review some of the nonlinear optics issues associated with this technique. We shall also examine several current applications, including probing electronic conduction in liquids and examining the nature of single-cycle pulses of polarized electromagnetic radiation.

An optically controllable nonlinearity

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Enhanced optical reorientation leads to a high optical nonlinearity in dye doped nematic liquid crystals (NLCs) [1]. We show that in Azo dye doped nematics, the magnitude and even the sign of the optical nonlinearity can be controlled by light.

Azo dyes have two conformational states, an elongated trans isomer and a bent cis isomer. In a nematic host, these two isomers cause independent nonlinearities, similarly to two different non-isomerizable dye dopants. Consequently, the nonlinearity of an LC host containing a mixture of both trans and cis isomers depends on the equilibrium distribution of the two isomers, which can be adjusted by photoisomerization with a control beam. The resulting controllable nonlinearity may be used for all-optical switching or light addressable displays.
Besides this unique property of Azo dye doped LCs, an understanding of the responsible mechanisms provides important information for related systems such as Azo dye doped alignment layers or Azo sidechain polymers, which are candidates for optical data storage.

A simplified model gives an idea about the origins of the enhancement of optical reorientation in dye doped LCs: Due to their dichroism, the excitation probability of the dye molecules depends on their orientation with respect to the electric field vector of the linearly polarized light wave. An additional torque is exerted on the director causing enhanced reorientation if the intermolecular interactions are different for the molecules in the ground and in the excited state. Besides state-dependent intermolecular interactions, state-dependent rotational diffusion can also cause the enhancement [2]. For small dye concentrations, the effect of the dye is proportional to its concentration.

Azo dyes can undergo photoinduced transitions between their trans and cis isomers. The isomers differ in properties such as their dichroism and absorption spectra, i.e., their excitation probabilities and thus the trans-cis equilibrium depend on the wavelength and on the light field components parallel and perpendicular to the director. Due to relaxation in the direction of the thermal equilibrium, the photoinduced trans-cis equilibrium is a saturation function of the intensity.

We present a phenomenological model of dye enhanced optical reorientation with consideration of photoisomerization. We ascribe an individual enhancement factor to the trans and the cis isomer, respectively. The concentration ratio of the isomers in turn determines the overall enhancement factor of the mixture.

In the experiments, an extraordinarily polarized pump beam induces reorientation and sets a certain trans-cis equilibrium. Under the given experimental conditions this equilibrium is saturated and thus independent of the pump intensity. However, it can be adjusted by the intensity of an additional ordinarily polarized control beam that will not contribute to the reorientation. Our experiments show that the magnitude and even the sign of the nonlinearity can be adjusted by the control beam. We determined the individual trans and cis enhancement factors for several Azo dyes. In all cases, the cis form has a positive enhancement factor, and the trans isomer has a negative one.

This is an important hint to a systematic mechanism of photoinduced reorientation in Azo dye doped LCs. It is essential for an improvement of the functionality and performance of the material to understand such responsible mechanisms.

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Phase only modulation by Polymer Dispersed Liquid Crystals

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The typical size of the liquid crystal domains in Polymer Dispersed Liquid Crystal (PDLC) is of the order of micrometers. As a consequence a strong scattering of light in the visible range is observed leading to an opaque state which can be switched to a transparent state by a suitable voltage. An increase of the efficiency of the phase separation process, which depends on the components concentration and UV curing parameters, may lead to formation of sub-micron (nano-sized) droplets [1][2]. In this work we report the method of preparation of a new class of nanometer-sized PDLC obtained by using a multifunctional vinyl monomer in a pre-polymer mixture which is suitable for fast polymerization process. A systematic analysis of the sample morphology and electro-optical characterisation has been performed on a large number of samples. These samples have high transparency to the visible light. However, reorientation of the liquid crystal domains due to the application of a suitable voltage can affect the optical phase shift.

Measurements have been performed by using an interferometric technique and phase shifts over $\pi/4$ have been detected with an applied effective field of about 20 V/μm [3]. These materials appears suitable as polarization independent "phase only modulators" to be considered for telecom applications.

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