Nonlinear optics in daily life

Elsa Garmire[*]

Thayer School of Engineering, Dartmouth College, 14 Engineering Drive, Hanover, NH, 03755, USA
[*]*garmire@dartmouth.edu*

Abstract: An overview is presented of the impact of NLO on today's daily life. While NLO researchers have promised many applications, only a few have changed our lives so far. This paper categorizes applications of NLO into three areas: improving lasers, interaction with materials, and information technology. NLO provides: coherent light of different wavelengths; multi-photon absorption for plasma-materials interaction; advanced spectroscopy and materials analysis; and applications to communications and sensors. Applications in information processing and storage seem less mature.

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1. Introduction

Today, after more than fifty years of development, the laser is used in innumerable every-day applications. Nonlinear optics (NLO) is only a year younger than lasers as a research field. Comparing Google website statistics, "Applications of Nonlinear Optics" has 5% as many websites as "Applications of Lasers" (10^6 compared to 18 x 10^6). Refereed papers, through Web of Knowledge, show similar statistics (4%). Some might take this to mean that nonlinear optics has only 5% the impact on life as lasers. This paper will show, however, that a large number of laser applications owe their existence to nonlinear optics, if not always explicitly stated.

The impact of NLO on science is widely understood (at least by those in the field) and has enabled, in one way or another, at least nine Nobel prizes in physics and chemistry. NLO phenomena have been observed at wavelengths from deep infrared to extreme UV, and even used to generate THz radiation. Optical nonlinearities are exhibited by crystals, amorphous materials, polymers, liquid crystals, semiconductors, organics, liquids, gases and plasmas.

Early applications of NLO included second harmonic generation, Q-switching and mode-locking, all of which extended the applications of lasers. Fiber optic communications early showed the deleterious impact of NLO in glass. With narrow-line lasers, ultra-fine resolution spectroscopy became possible. Today we see a plethora of promising future applications for NLO, including quantum optics, quantum computing, ultra-cold atoms, plasma physics and particle accelerators, to name a few, with many more to come.

This paper focuses on only those applications that impact daily life today; enumerating its impact on science as well would have been a massive undertaking. In an overview of NLO and its applications [1], the author has considered χ_2 nonlinearities: harmonic generation and frequency mixing; optical parametric oscillators. The χ_3 nonlinear refractive index yields self-phase modulation, mode-locking, wave-mixing, photorefractivity, phase conjugation, spatial solitons, nonlinear waveguides and interfaces, and optical bistability. Nonlinear absorption includes multi-photon absorption, saturable absorption, multi-photon filament formation, optical limiting and Q-switching, among other applications. Nonlinear optical scattering processes are predominantly stimulated Raman and Brillouin scattering. From this vast array of NLO effects, which seemed to offer much promise, only a few have made it to commercial reality. This paper assumes familiarity with nonlinear phenomena; if not, the web offers details on each nonlinearity mentioned here. As well, the author has published a resource letter for references in each of these areas [2].

This paper will classify applications as shown in Fig. 1. At the highest level, NLO has had a profound effect on lasers, laser-materials interactions, and on the general field of information technology, which includes sensing, communicating, storing and processing of information. Each of these areas is discussed in a separate section below. In the end, the impact of NLO on daily life must also include all applications that use lasers that have been diversified by means of NLO.

2. Diversifying lasers and coherent light

The simplest lasers have a time dependence determined by their pump; their wavelength is determined by their gain medium. Inserting a NLO element within the laser cavity makes possible a diversity of new lasers and applications. Alternatively, the NLO element may be located outside of the laser cavity, thereby diversifying the character of the coherent light emitted by the laser. The NLO process may alter the color of the laser light, and/or create pulses of laser light. A third application is the control of the spatial and/or frequency characteristics of the laser output through NLO methods.

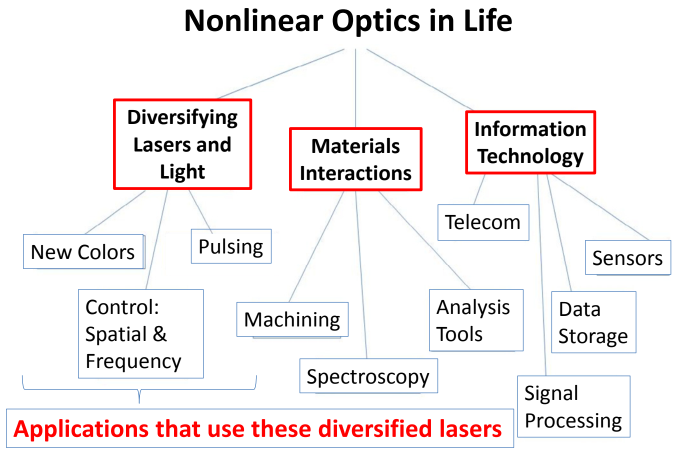

Fig. 1. Scheme for organizing nonlinear optical applications.

For many applications that use NLO today, competing approaches may develop that do not use NLO. While NLO might be favored now, it is not clear which technology would be better in the future. Thus this paper is a snapshot only of today's technology.

2.1 New colors

Direct generation of blue/green lasers has traditionally been very inefficient (e.g. the argon laser). To reach short wavelength coherent light, it is generally much more efficient to generate second and third harmonics of infrared lasers. Today, however, strong competition is coming from blue and UV laser diodes. Only time will tell which technology will be used for which applications. The extension of multiple harmonics into the EUV has exciting future potential, such as for very high resolution lithography and accelerating elementary particles to very high energy, but since EUV is not yet in commercial application, it will not be discussed here.

2.1.1 Harmonic generation

Second harmonic generation (SHG) of the Nd:YAG laser (wavelength = 1.06 μm) and its solid-state laser cousins have been the most efficient source of coherent green light (wavelength = 532 nm) for a number of years. The ubiquitous green laser pointer is the culmination of applying NLO to efficiently turn invisible Nd:YAG output into highly visible green. As Fig. 2 shows, the NLO crystal KTP is the largest active component in the green laser pointer because all optical nonlinearities are weak. Even so, harmonic generation is a lossless process and diode-pumped solid state lasers (DPSS) can be very efficient. When the nonlinear crystal is placed inside the laser cavity, the efficiency of second harmonic can approach 80%. The larger optical fields of picosecond (ps) or femtosecond (fs) pulses can generate second harmonic even outside the cavity with efficiencies approaching 100%.

Efficient generation of short-wavelength coherent light opens up a wide variety of photochemical reactions that are valuable because they proceed differently than thermal reactions. Photocatalysis, photodissociation, photoelectrolysis, photosynthesis, photo-polymerization, photoresist (for lithography), are all processes that require short-wavelength light. These processes use SHG of Nd-based lasers for localized processing and/or specialized excitation with ps pulses. While these processes may not directly require NLO, they do require short wavelengths that are often fulfilled most conveniently by SHG.

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The Ti:sapphire laser is a uniquely important solid-state laser with a broad gain line that can be mode-locked to provide fs pulses. Its absorption band peaks around 450 – 500 nm and can be optically pumped with frequency-doubled Nd:YAG lasers. An example is Coherent's Mira Ti-sapphire laser, pumped by its Verdi DPSS 532 nm laser [4]. However, as blue laser diodes become more powerful and robust, they may replace the SHG of DPSS lasers for many applications. For example, one company is already developing direct blue-diode pumped titanium sapphire lasers [5].

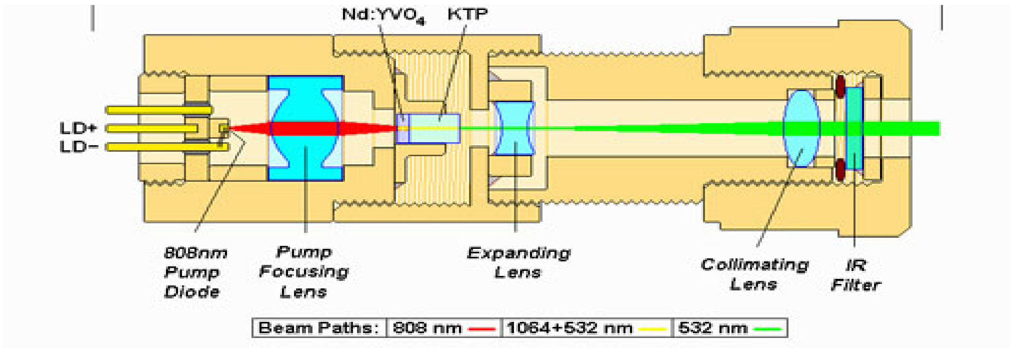

Fig. 2. Diagram of a green Diode-pumped solid state laser pointer [3].

2.1.2 Optical parametric oscillators

Practical optical parametric oscillators (OPOs) have come of age with the advent of ultrashort pulses for high peak power, new and effective nonlinear crystals, and periodic poling for phase-matching. Robust, turn-key and/or computer-operated, OPO's are now widely used for spectroscopy, as pointed out by Continuum, whose integrated system, with a Q-switched YAG laser as pump is "a research tool that will transform the way spectroscopists work. Instead of multiple complicated devices, scientists have one all-encompassing tool allowing them to measure the whole spectrum" [6]. Wide spectral range is available by pumping with second and third harmonics as well as the fundamental of the diode-pumped Nd:YAG laser whose pulses are in the ns range.

OPO's are available from practically all the major laser-makers. Advanced designs with ultrafast pumps enable a tuning range from 345 nm to 2.5 μm gap-free with no change of intracavity optics or crystals. The fs OPO is synchronously pumped with a mode-locked Ti:Sapphire laser with a pulse width adjustable from 80 to 350 fs [7].

Continuous OPO's operating in the mid-IR are common. One system is all-in-one, with a diode-pumped Nd:YAG laser and a PPLN:Mg (periodically poled, MgO doped lithium niobate) parametric crystal. This compact, robust all-solid-state OPO is available for laser spectroscopy over a large tuning range in the mid-infrared. Coarse tuning is done by manually changing the lateral position of the PPLN crystal to select a suitable grating spacing, with 100 mW maximum output for 1.2 W of laser pump power [8]. For higher CW powers, a 15 W Nd-doped fiber laser at 1.06 μm wavelength pumping an OPO using PPLN:Mg can generate mid-IR frequencies with up to 70% conversion efficiency [9].

2.1.3 THz generation

An OPO that generates THz is available commercially [10]. More commonly, broad-band terahertz radiation can be generated by means of optical rectification from fs pulses that rapidly change in time. With fs pulses, the "static" field is time-varying and radiates broadband free-space waves at THz frequencies. The χ_2 materials are typically ZnTe and GaAs. However, a strongly competitive technology uses the linear electro-optic effect; in the future, THz competition may also come from quantum cascade lasers. Today, few THz systems have reached the stage of "practical," although the technology is rapidly improving.

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2.1.4 Stimulated Raman lasers

Stimulated Raman scattering (SRS) offers gain at frequencies separated from the laser pump by a molecular vibration. The variety of laser pump wavelengths and SRS materials provides exceptional diversity for lasers. The most important application is Raman lasers and amplifiers in fibers for telecommunications, discussed later. In general, bulk Raman lasers require high-power pulsed laser pumps and provide an output at longer wavelengths. Geometries may be single pass amplifiers, resonators (Raman lasers), oscillator-amplifier configurations and/or waveguides. SRS generation can be intra-cavity or extra-cavity. Long fiber lengths enable low Raman laser thresholds.

Raman lasers have the greatest value when they generate wavelengths not available by other means. Raman lasers make eye-safe LIDAR (light detection and ranging) by shifting the 1.06 μm Nd ion lasers to safer longer wavelengths. Raman fiber lasers offer the highest CW output power of any source in the eye-safe infrared: 300 W with 85% efficiency [11]. A yellow 589-nm Raman laser can be used for a laser guide star (sodium beacon for atmospheric correction) in astronomy when combined with adaptive optics. The yellow wavelength can be achieved by pumping a barium tungstate ($BaWO_4$) Raman emitter placed inside a Q-switched Nd:YAG laser; the Stokes output is at 1180 nm and is then frequency-doubled [12]. Medical lasers also want yellow/orange wavelengths, not available from other lasers. The Raman laser microprobe will be discussed later. Diamond has also produced a Raman laser. It remains to be seen how many of the different Raman laser types will be commercially practical.

The largest Raman shifts come from pressurized hydrogen and methane, which have the advantage that, as gases, phasematching is not a major problem. In gases there's no self-focusing, Brillouin scattering or anti-Stokes to worry about (unlike in solids). However, pressurized gases are not always easy to work with. These are used in specialized high-power applications only.

One last (future) Raman laser application may be in silicon, which does not emit light from band-to-band recombination, and so has been unable to be used in chip-to-chip communication within computers. However, SRS can turn silicon into an emitter, when pumped external to the silicon chip. While Intel experiments reported an impractical 180 mW threshold for a CW silicon Raman laser [13], Japanese researchers have recently reported μW-threshold Raman lasers in silicon with μm-scale geometry [14]. It remains to be seen if this will become useful.

2.1.5 White light continuum

One last source of new wavelengths comes from self-phase modulation and four-wave mixing (FWM) of ultra-short optical pulses in fibers (or water). Picosecond pulses in water or femtosecond laser pulses transmitted down fibers are frequency-broadened by these nonlinear processes into a white-light spectrum. This white-light source, covering the entire visible range, down into the infrared and even into the UV, is particularly valuable for ultra-fast excite-probe spectroscopy.

2.2. NLO for pulsing lasers

Nonlinear optics can create short laser pulses either by Q-switching or mode-locking. Traditional laser cavities contained a rod-shaped crystal along with a nonlinear element (or nonlinear mirror). More recently, diode-pumped microchip lasers have been shown to provide compact, robust sources with a nonlinear Q-switch element to provide high instantaneous power, enough to efficiently generate second harmonic in a phase-matched χ_2 crystal.

2.2.1 Q-switching

Q-switching provides the highest pulse energies, usually pulses typically ns long. Passive Q-switching relies on nonlinear absorption and/or nonlinear refractive index. These nonlinear elements offer a simple, robust means to obtain 10^6 times or more peak power than CW lasers. Commercial passive Q-switched microchip lasers (Nd:YAG, for example) can generates pulses as short as 100 ps with a semiconductor saturable absorbing mirror (SESAM), or pulses ~1 ns long with a Cr^{2+}:YAG saturable absorber. Such NLO microchip Q-switch lasers can have very high pulse repetition rates: 100 kHz, up to MHz. Their pulse energies are on the order of a few mJ, yielding peak powers of many kW.

Alternatively, the gain medium can consist of a diode-pumped fiber doped with rare earth with a passive Q-switch in the cavity. The Nd:doped fiber laser emits up to 10 mJ energy per pulse @ 20 kHz, with up to 200 W average output power and 400 ns pulse duration for 20 kW peak power [15]. High power fiber lasers in the eye-safe wavelength region, 1.5 – 2.1 μm, such as Er, Tm, or Ho fiber lasers, can use Cr-doped ZnSe as their Q-switch. These and similar high power fiber lasers and amplifiers are revolutionizing the performance and utility of high power lasers.

For the highest power Q-switched lasers, DPSS geometries are specially designed to optimize lateral pumping and intra-cavity harmonic generators, leading to peak powers as much as 180 kW, repetitively pulsed at 5 kHz [16]. Applications for these and other lower-power Q-switched lasers include materials processing: welding, cutting, micromachining, marking/identification, sintering, brazing, soldering and altering material phases. Lower powers are sufficient for most micromachining, telecommunications, medical and other advanced applications.

The competition to passive Q-switching is active Q-switching with electro-optics or acousto-optics. Active Q-switches offers the opportunity to time pulses according to some external stimulus, but their system requires a more complex design.

2.2.2 Mode-locking

Locking all the longitudinal modes in a highly multimode laser produces much shorter pulses – from several ps to fs, depending on the bandwidth of the gain medium. This can be done by NLO using saturable absorbers that produce sub-ps pulses from Nd-doped glass and ~10 ps pulses from Nd:YAG. SESAMs create fs pulses from Ti:sapphire lasers; an alternative nonlinear mechanism for these lasers is Kerr-lens mode-locking. The competition to passive NLO mode-locking is active acousto-optic mode-lockers, although this requires tight frequency control.

2.2.3 Solitons

Another commercial pulsed laser using NLO is the soliton laser, which has the advantage of ultra-stable operation – high repeatability with clean transform-limited pedestal-free pulses. A practical fiber-based fs soliton laser operates at 1.5 μm, the important wavelength for telecommunications [17].

2.2.4 Measuring femtosecond laser pulses

Femtosecond lasers could not have been developed without a way to characterize the pulses they emit. Instrumentation that measures both the amplitude and phase of fs pulses requires NLO. The autocorrelator has been key, not only to the development of fs technology, but also to the on-going characterization of fs pulses [18]. The length of the pulse overlap in physical space determines its length in time. Pulse-overlap monitoring requires a NLO process (usually SHG) to separate true pulse length from simple interferometry, which only measures spectral width. Without NLO, the entire field of ultra-fast optics would be severely hampered.

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2.3 Spatial and frequency control of lasers

The NLO process of phase conjugation has been used for laser beam clean-up, reducing both spatial and frequency modes. The Stokes light retro-reflected by stimulated Brillouin scattering (SBS) within a fiber will remove aberrations and be diffraction-limited. SBS is used to clean up the spatial and frequency of the pump laser. SBS may limit the power achievable through a single fiber (as discussed later), but it also provides a means of coherently combining multiple beams. SBS locks the phases from parallel amplifiers by generating a phase conjugate reflection that propagates back through the amplifiers in a second pass, reconstructing the initial phase profile.

The Brillouin laser is a highly coherent light source. One example demonstrates a 20 dB reduction of RIN and frequency noise compared to the narrow-linewidth Er-doped fiber laser pump source [19]. Stable operation requires active stabilization to lock resonance between the pump laser frequency and the Brillouin cavity mode.

3. Laser-materials interactions

In the previous set of applications, most of the nonlinearities resulted from light interacting virtually with the material. This section considers applications where the light changes the material through which it passes, either permanently or transiently. These applications are labeled as: machining (where material is changed permanently), spectroscopy (where atoms and molecules are changed temporarily) and analysis tools (where selective species are changed). The latter comprises a wide variety of applications where selectivity in the nonlinear process enables the material to be analyzed.

3.1 NLO for permanent changes in materials

Lasers can permanently alter materials (change phase, remove or add material, etc.) in two ways: either through heat or through NLO processes. The latter is of interest here and requires short enough laser pulses that heat generation is kept to a minimum. Traditionally, ns pulses have been used, although more recently ps and fs pulses have become particularly useful.

The NLO process usually involves multiphoton absorption that leads to ionization at the focus of the laser beam, which creates a localized plasma (a spark). The plasma vaporizes the target, removing material (ablation). A single pulse will dig a very small hole into the sample; repetitive pulses can drill holes or, with sample motion, can cut lines that engrave a pattern in the surface. With computer control and continued processing, highly accurate laser machining can be performed. Because multi-photon ionization has a threshold, holes can be smaller than the diffraction limit. The entire basis of high quality laser machining, then, relies on the nonlinear process of multiphoton absorption.

In laser machining, once a plasma is generated by multiphoton ionization, it continues to absorb more light and sustains itself until the pulse is over. Once multiphoton ionization threshold is reached, the plasma absorption is independent of the material; thus this process even works on reflective surfaces (destroying their reflectivity).

The formation of a spark also introduces a surface shock wave that can be important in the ablation process. The shock wave can help in laser desorption, a process by which the laser removes particles (such as dust) attached to the surface. Ablation of thin films is used extensively in the semiconductor industry for removing conducting ITO electrodes on solar cell edges, trimming resistors, removing short circuits, etc. The industry uses laser machining for die cutting and marking, as well as printed circuit board drilling and structuring, and marking LED chips.

The material ablated during the multiphoton ionization process may be deposited on a separate substrate placed nearby, forming its own thin film, in a process similar to e-beam or ion-beam sputtering but is much cleaner. Photo-ablation has been used to create films of nano-materials, superconducting materials, thin dielectrics, oxides, nitrides, metals and carbon based materials. A later section will describe how biological samples are analyzed by multi-photon ionization.

Recently, ps and fs pulses have been used for laser machining because, with ns pulses, some of the material may sputter or splash out, leaving a rough edge. With ps or fs pulses, the ablation and hole-digging process can be very clean. The ultrashort pulse duration eliminates thermal effects that cause microcracks and molten debris. The high peak power can machine copper, glass and ceramics, which are often difficult for ordinary laser machining. Because the process is highly nonlinear and does not depend on linear heat absorption, it works on metals and dielectrics, thin films and even inside transparent materials. Small, compact fs lasers can be made with low electrical consumption and high reliability for operation in industrial environments, often based on diode-pumped fiber lasers.

Micromachining with fs pulses can create very fine structures, such as Micro-Electrical-Mechanical Structures (MEMS) in silicon. Laser micromachining affects daily life through medical device manufacturing. Implants and surgical tools require advanced manufacturing processes because small changes in surface texture, dimensions and shape can affect their performance. Ultrafast lasers can process virtually any material without heat dissipation, and therefore with an extremely high accuracy and quality. A new generation of coronary stents, finely processed catheters and surface textured bio-implants have all reached the market through laser micromachining (see Fig. 3). Highly accurate microfabrication is fulfilled by computer-guided laser ablation of unwanted material. With a fs fiber laser, for example, a 15 μm cut width can be made in materials with 100μm-wall thickness [20]. New applications for micro-machining are being actively developed today, using either nano-structuring or advanced additive manufacturing.

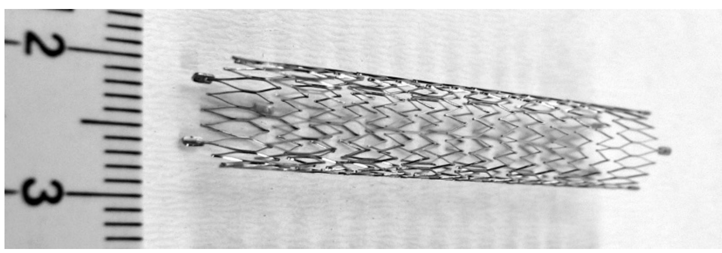

Fig. 3. Biomedical stent: Adapted from http://en.wikipedia.org/wiki/Stent.

Microsurgery, performed by the multiphoton ionization of biological tissue by fs laser pulses, has all the advantages of micromachining. Precise ablation and cutting is possible at the subcellular level. Using a focused fs laser, localized multiphoton excitation can open a nanoscopic pore in a single cell membrane to allow transfection (introduction of nucleic acids). Localized photochemistry is also possible, offering, for example, 3D photolysis of molecules in femtoliter volumes.

Femtosecond LASIK is replacing standard LASIK, uses excimer lasers, because of better performance: more accurate cutting and less damage. The third harmonic of Nd-based lasers at 355 nm is usually used because UV wavelengths are more effective in materials transparent in the visible. Third harmonic of Nd:YAG lasers is also used for marking glass surfaces, as well as inside glass, either at the focal point of a lens or where two beams are made to overlap at localized points where small bubbles can form.

This last process occurs in the “laser crystals” that are readily available as novelties and souvenirs – a demonstration visible to anyone of the power of NLO laser-materials transformation. Figure 4 shows a “laser crystal,” with an image formed by nonlinear absorption, plasma and bubble formation, manufactured in Russia in 1989. This very early example used a computer to control the lateral location of the focal point of a laser while the sample was turned on a lathe. Today two-axis computer control enables any kind of 3D image to be created inside a glass block, with fs lasers providing much higher resolution.

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Fig. 4. Laser "crystal" photograph by the author.

UV lasers also perform additive manufacture by means of threshold polymerization processes. When traditional laser stereolithography and 3D printing use processes with thresholds, then irradiation with two overlapped beams can harden a specific location within a liquid, yielding a 3D object. The two-photon polymerization process is most common, but nonlinear absorption can also be used for additive manufacture.

3.2 Advanced spectroscopy

Within a year of the first gas laser, saturation-dip spectroscopy had been developed. Two counter-propagating waves saturate the Doppler-broadened gain profile for the velocity packet of molecules at line center. The resulting Lamb dip exposes the natural linewidth, which enables high-resolution spectroscopy (and frequency stabilization of lasers). Saturation spectroscopy can reveal hyperfine structure due to spin-rotational interaction with a resolution of one part in 10^{11}. Two-photon absorption spectroscopy can provide similar Doppler-free resolution without requiring gain.

Stimulated Raman Scattering can be used to increase the Raman signal intensity in Raman spectroscopy. Most common is Coherent Anti-Stokes Raman Spectroscopy (CARS), in which two intense collinear laser beams irradiate a sample. One laser is held at constant frequency, while the other is tunable. When the frequency difference coincides with a Raman active mode, the Raman signal will be enhanced by typically 10^5. While a tunable laser is required, a spectrometer is not needed.

Nonlinear Spectroscopy uses an incident laser light field to perturb the optical properties of a molecule (or atom). Subsequent light probes see changes in molecular states. Experiments typically involve SHG or FWM. These processes depend on the spatial and temporal overlap of optical pulses. With pulses shorter than dephasing and relaxation times, probing with a pulse at a known time after the excitation pulse can elucidate how molecular states evolve in time.

Optical biopsy is an application for nonlinear spectroscopy, such as distinguishing cancer cells from normal cells. This relies on the observation that cancer cells have different relaxation times from normal cells. Measurement of the time decay of second harmonic on a biopsy sample can separate cancerous from normal cells. This is only one of a myriad of medical applications that are based on nonlinear spectroscopy.

Ultra-fast absorption spectra are measured by generating a white light continuum, already described. The white light pulse probes the absorption spectra after a first excitation pulse. As time delays are added, transient spectra can be recorded.

In FWM spectroscopy, two pump waves interfere to introduce a nonlinear grating; a probe wave diffracts from this grating to form a fourth "signal wave." FWM provides background-free detection, since the signal wave appears at a different angle from the rest of the light. The diffracted signal intensity can be measured as a function of time, applied electric field, writing beam intensities, etc. Transients are measured with pulsed pump and time-delayed probe,

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determining excited state lifetimes, dephasing lifetimes, as well as diffusion and drift in semiconductors.

3.3 Analytical tools

Nonlinear spectroscopy, already discussed, is often used for materials analysis. Other nonlinear techniques have become very important. The first is the analysis of atoms and molecules released by multi-photon ionization during the process of laser ablation or desorption. The second is nonlinear microscopy, in which a nonlinear process separates one part of a sample from the rest, or from background.

3.3.1 Laser induced breakdown spectroscopy (LIBS)

In LIBS, a short laser pulse creates a micro-plasma on the sample surface and spectral features of these ions are identified by traditional spectroscopy. Chemical analysis of a single spot can be made extremely fast, with measurement times on the order of a few seconds. LIBS is good at identifying lighter elements, such as H, Be, Li, C, N, O, Na, and Mg.

Pulsed infrared lasers in biological samples have large penetration depth and can remove large quantities of material with each laser shot. Biomolecules can be ionized and vaporized by laser irradiation of polyacrylamide gels, tissue sections, and bacterial cells. In general, ionization by an intense laser pulse on a macromolecule causes cleavage into tiny fragments and the loss of its structure. In 2002, Koichi Tanaka won the Nobel Prize in Chemistry for the discovery that when immersed in a matrix consisting of ultra-fine metal powder in glycerol, a macromolecule can be ionized with a laser pulse without losing its structure. This technique, initially known as soft laser desorption ionization (SLDI), is now more regularly referred to as MALDI (matrix-assisted laser desorption ionization). The organic matrix efficiently absorbs the laser energy so that upon exposure to pulsed laser radiation (usually UV), the ensuing phase transition of the matrix volatilizes the large molecules. A small fraction of these molecules are also ionized, so that a mass spectrometer can accurately determine their mass. The combination of MALDI with mass spectrometry is an enabling technology for the emerging discipline of proteomics. A fascinating instance of MALDI in the real world is the analysis of the clothing of “ice man,” the Tyrolean mummy, Oetzi [21].

3.3.2 Imaging nonlinear processes

Combining microscopy with NLO changes in materials can be particularly valuable. Using confocal microscopy, biological systems and processes can be analyzed *in vivo* at the microscopic level. NLO offers higher resolution images and localized functionality. Many of the NLO processes discussed above can be combined with imaging through a confocal microscope: SHG, THG, CARS, and SRS. Because it requires phase-matching, SHG can give additional information on structural data, by separating out different components in tissue without specific staining. SRS can identify microscopic local changes in chemical composition.

Particularly important is two-photon excited fluorescence (TPEF), which involves two-photon absorption of dyes followed by fluorescence. These studies can provide structural information deep within tissues and/or living organisms, as well as isolated excitation within a localized, confocal volume. This is because the laser intensity drops rapidly outside focal plane, ensuring no tissue damage caused by photo-toxicity. A single pulsed laser of wavelength 700 to 1300 nm can excite two or more fluorescent dyes. Two-photon microscopy is commercially available from Olympus, Leica and Zeiss, to name a few companies, and may someday be in use in clinical settings [22,23].

Analysis of localized photochemical reactions can provide even more information. For example, bimolecular interactions can be detected through multiphoton two-color cross-correlation spectroscopy. Multiphoton fluorescence correlation spectroscopy provides measure of diffusion, while fluorescence photobleaching recovery time offers additional information.

As a commercial example, a SRS spectroscopy system has been developed to provide a real-time (less than one second) automated identification of skin cancer through observing spectral changes associated with the biochemistry of skin cancer cells [24].

4. Information technology

The dominant information technology impacted by NLO is fiber optic telecommunications. As the use of optical sensors is rapidly developing, NLO shows up in various ways; today, however, it appears that major inroads have not yet been made by NLO technology. As far as signal processing and data storage are concerned, many suggestions have been made, but very few have been commercialized.

4.1 NLO in telecommunications

Soon after fiber optic telecommunications were introduced, NLO was found to be the fundamental limit to the amount of data that can be transmitted on a single optical fiber. As laser power levels increase, NLO limits data rates, transmission lengths, and the number of wavelengths that can be simultaneously transmitted. NLO showed up first in undersea installations, where fiber lengths were the longest. It is now known that NLO must be considered whenever designing state-of-the-art fiber optic systems. After transmission systems have been designed to overcome basic linear attenuation and dispersion, then NLO becomes important.

An upper limit on the optical power that can be usefully launched into an optical fiber is due to stimulated Brillouin scattering (SBS). When the SBS threshold is exceeded, some of the light in the fiber is reflected back to the transmitter. SBS thus saturates the optical power that can be received and SBS back-reflections cause noise that degrades system performance. The deleterious effects of SBS can be reduced by using laser pulses that have a spread of wavelengths, thereby reducing the Brillouin gain.

Stimulated Raman scattering (SRS) in telecommunication fibers is a much smaller problem than SBS because of high SRS threshold. The SRS threshold is close to 1 Watt in a typical fiber, which is a thousand times larger than the SBS threshold. However, systems are already deployed that use erbium-doped fiber amplifiers (EDFA) with high power pump lasers to boost optical signals along the fiber. The EDFA pump power is at 980 nm and first Stokes SRS will shift the wavelength to 1.55 μm where it can interfere with communication signals. In high power wavelength division multiplex (WDM) systems, the effect of SRS is to rob shorter wavelength channels of power as the SRS Stokes process converts the power to the longer wavelength channels.

On the positive side, Raman fiber amplifiers (RFA) and Raman lasers have proven to be practical sources. Plain silica fiber (unlike erbium-doped fiber) provides gain through the nonlinear process of SRS. In a variety of applications SRS amplifiers can replace EDFAs. Typically a wavelength of 1480 nm is chosen for the pump, which amplifies a signal at 1570 nm wavelength. RFA's have less noise and a very wide bandwidth, as shown in Fig. 5; they are becoming mainstream additions to long-haul telecommunication systems.

With optical feedback, RFA's become Raman fiber lasers, recently developed to provide new wavelength sources for a variety of applications. Raman fiber lasers are highly reliable optical sources at telecommunication wavelengths, from 1100 to 1700 nm, with CW output powers approaching ten watts. High-power pump diode lasers are not so sensitive to temperature variations as communication wavelength laser diodes and do not need thermo-electric cooling. Raman fiber lasers can cover a wide range of wavelengths with good stability, particularly valuable for Dense Wavelength Division Multiplexed (DWDM) transmission in ultra-long haul systems. They can be introduced to glass fibers already

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installed, providing distributed ultra-wide-band Raman amplification, leading to repeaterless submarine and very long span systems.

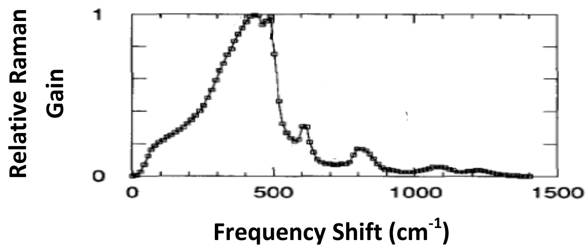

Fig. 5. Raman gain spectrum in a telecommunication fiber, adapted from Liu and Garmire [25].

Four-wave mixing has become a challenge in telecommunication systems that use Wavelength Division Multiplexing (WDM), a technology that puts many closely-spaced wavelengths on the same optical fiber. When the wavelengths have equal spacing, FWM causes cross-talk and distortion because the nonlinear refractive index creates sidebands separated by the difference frequency. These sidebands interfere with the original signal and produce new frequencies. The FWM process increases with smaller channel spacing and decreases with increased dispersion. Advanced system designers are well-aware of these NLO effects and take them into account in their designs.

Four-wave mixing can also be a useful tool in telecommunications. WDM systems, with many wavelengths on the same optical fiber, often need to convert from one wavelength to another. FWM provides parametric wavelength conversion through difference frequency generation. This process is instantaneous, simultaneously converting multiple channels up and down with equal efficiencies. The NLO process produces negligible spontaneous emission noise and has no intrinsic frequency chirp.

Solitons, pulses with a unique shape in time that travel through a medium without changing, require a balance between nonlinearity and dispersion. In fibers, self-phase modulation chirp is balanced by group velocity dispersion chirp. Are solitions used in telecommunication systems? The answer depends, to some extent at least, on definitions. Long-haul telecommunication systems are ultimately dominated by loss and must include amplifiers. When solitons are re-defined to include gain as well as loss, then it may be that some of these systems do, indeed, operate with pulses that could be defined as solitions. It is not clear, however, that they are solitary waves that form only for discrete values of the pulse energy.

4.2 NLO in data storage

Ordinary interferometry is generally a linear process. Data storage by holography would not, then, be a process that NLO can claim for itself. Compact disk masters were initially made with an argon laser, and the holes were created thermally. Again, not a nonlinear process. The holes in today's high resolution DVD's, however, require much a higher resolution manufacturing process, undoubtedly using the multi-photon ionization microfabrication technologies described in section 3. Thus NLO can claim an impact on DVD's.

The impact of NLO may eventually be seen with storage of data in three dimensions (rather than the 2D of CD's). Efforts to store data using the photo-refractive effect in lithium niobate have not achieved commercial success. Research continues on 3D digital data storage using other NLO processes, such as spectral hole burning, which unfortunately requires cryogenic temperatures. Several companies are actively developing the technology, but none is available at the present time.

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In recent years, 3D video images have become increasingly popular. A role for NLO appears to be in real-time 3D image formation. University of Arizona has shown that 3D holographic images can be recorded using polymer films made photorefractive by applying a lateral field and relying on a nonlinear refractive index and/or absorption of a chromophore. A new doped-polymer material has been developed for a holographic 3D display capable of refreshing images every two seconds, thereby demonstrating telepresence [26]. Proposed applications include telemedicine, prototyping, advertising, updatable 3D maps and entertainment. It will probably be a while, however, before these early-stage systems will be seen in commercial markets.

4.3 NLO sensors

Most optical sensors (laser radar, intrusion detection, temperature sensors, etc.) do not require NLO. However, many of today's optical sensors are based on optical fibers, where NLO may begin to play a role. Brillouin fiber sensors are sensitive to a number of variables, and Raman fiber sensors can measure temperature. It is unclear, however, if any of these sensors actually use nonlinear properties of the sensing material.

A variety of nonlinear sensors seem to be coming on the market, but very often the technical details of these sensors are proprietary. While they may not overtly use NLO, they may use NLO in their manufacturing processes. For example, many sensors use Bragg mirrors written within the fiber, a process that most likely involves NLO.

5. Conclusions

NLO provides major benefits (as well as some limitations) to some very important applications in: commercial lasers, telecommunications, sensors, environmental monitoring, medicine, manufacturing and materials processing, the military, and in scientific instrumentation. Taken together, these applications have considerable impact on daily life.

Arguably, NLO has had its greatest impact in advancing science. The numerous successful applications to science were not included here; that would take another paper at least. This paper mostly avoided applications that have not yet made it to the marketplace. Over the years, NLO has provided a stream of possible applications that have not yet come to pass. One example is optical signal processing, where past efforts have not led to practicality, although the future seems bright for quantum computing and quantum interference.

This paper focused on today's uses of NLO in technology. NLO has made possible a diversity of advanced lasers and coherent sources, has empowered micromachining, offered high-resolution spectroscopy, new materials analysis tools, and high-capacity telecommunications. Daily life without NLO would be missing the high quality fiber optics systems that power the internet, the technology that enables advanced medical devices, the important laser-based analysis tools used for medical diagnosis, characterizing new materials, monitoring pollution in air and water … and green laser pointers.

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