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Nonlinear Materials for Information Processing and Communications

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Phil. Trans. R. Soc. Lond. A 1996 **354**, 745-756
doi: 10.1098/rsta.1996.0028

Nonlinear materials for information processing and communications

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The current state-of-the-art of nonlinear optical materials suitable for ultrafast all-optical applications is discussed. This survey will include progress in the third-order nonlinear properties of semiconductors, polymers and glasses and examples of their applications to devices. Also discussed will be 'cascading', a second-order nonlinearity-based approach to all-optical effects.

1. Introduction

Data transmission rates for telecom have been doubling approximately every five years implying that > 5 Gbit s^{-1} systems will be operational by the year 2000. Non-telecom applications, such as virtual reality and remote medical diagnosis, require more than 100 Gbit s^{-1} information rates as soon as possible. Although electro-optic modulators have been successfully operated out to 50 Ghz, it is not clear whether this value can be reached (or even exceeded) outside a research laboratory environment. An alternative approach to switching information at very high speeds is to use optical interactions, in which signal beams can be controlled by either their own intensity or by their temporal overlap with other (control) optical beams. In this case, nonlinear materials and interactions are required, the optical properties of which can be changed by the presence of strong enough optical beams. The intensity dependence of the refractive index, a third-order nonlinear effect, has received most attention. In addition to addressing the state-of-the-art in such materials, here we will also consider other effects (cascading) which lead to a nonlinear phase distortion without necessarily introducing an index change in the usual sense.

Index changes can be induced via a number of different physical mechanisms in a material. Bearing in mind that we are primarily interested in materials which respond on sub-nanosecond time scales, the only two effects which turn out to be useful are Kerr nonlinearities, which result from the nonlinear response of bound electrons in their potential wells, and electron excitation from ground to excited states facilitated by the absorption of a photon. The first case is usually called the 'non-resonant' nonlinearity. The most frequently used example of the second case is carrier excitation in semiconductors. Figures of merit (FOM) have been defined for identifying suitable materials in both cases and these will be discussed here. In fact, for some years now, a major problem has been the lack of measurements of the parameters that are needed to evaluate these FOM: this continues to be a major problem. That is, there are many 'promising' materials reported every year, but such

Phil. Trans. R. Soc. Lond. A (1996) **354**, 745–756
Printed in Great Britain

labels are usually based on only one of the multiple parameters needed for identifying truly suitable materials.

We start in the following section by discussing the type of device functions of most interest to communications or signal processing. The requirements for a nonlinear material emerge from this device discussion. We then, in § 3, examine glasses, semiconductors and polymers as classes of potentially useful materials and the trade-offs between them. In § 4 we briefly discuss a different materials approach to all-optical phenomena, namely the cascading of second-order nonlinearities.

2. Devices and material figures of merit

Nonlinear optics can play many different roles in signal transmission and processing. For example, temporal solitons provide a viable signal form for long distance transmission and are discussed elsewhere in this special issue (Doran 1995; Mollenauer 1995). Their very special properties are a direct consequence of nonlinear optics in silica-based fibres, which allows both soliton formation and transmission in the communications bands. Of principal interest in this paper are applications which involve the manipulation of signals, either in soliton or NRZ (non-return-to-zero) form (Agrawal 1989), i.e. methods to perform all-optical logic; all-optically controlled routing functions, such as multiplexing and demultiplexing of data streams; and, most recently, wavelength shifting. These operations are currently being pursued for high bit rate communications. However, it is important to note that there are other applications such as distributed networks for computing etc., image transmission for real-time medical diagnosis and entertainment (e.g. virtual reality), which will require data transmission and manipulation rates in excess of standard telecom rates and over shorter distances than required for commercial telecom. Although the transmission medium will almost certainly be fibre, data manipulation modulation etc. will probably require efficient nonlinear media.

Most all-optical devices are interferometric in nature and involve a control beam which changes the optical properties of the medium, and a signal beam which is affected by this change. The output signal can be displaced in space, time or wavelength, as shown in figure 1. The successful operation of such devices typically requires some minimum all-optically induced phase shift, $\Delta\phi^{\mathrm{NL}} = \Delta n(I) k_{\mathrm{vac}} L$ for a device of length L (Stegeman & Miller 1993). This phase shift can either be instantaneous and present only during the presence of the control pulse, or it can accumulate over the duration of the control pulse and then decay in time on a time scale that is long compared with the arrival and duration of the signal pulse. For an instantaneous Kerr nonlinearity, an index change of $\Delta n(t) = n_2 I(t)$ is induced by a local intensity I. In the case of an 'integrating' nonlinearity and control pulses much shorter than the relaxation time t,

$$\Delta n(t) = K \int \alpha I(t') \, \mathrm{d}t' \mathrm{e}^{-t/\tau},$$

where α is the absorption coefficient and K is a material constant.

The FOM quantify material limitations on the maximum achievable nonlinear phase shift. The effective device length can be limited by fabrication trade-offs, by attenuation due to scattering or by the absorption of one, two or more photons (Stegeman & Miller 1993). Beam attenuation is especially important for a nonlinear phase shift because $\Delta\phi^{\mathrm{NL}}$ depends on the intensity which decays with distance. An obviously

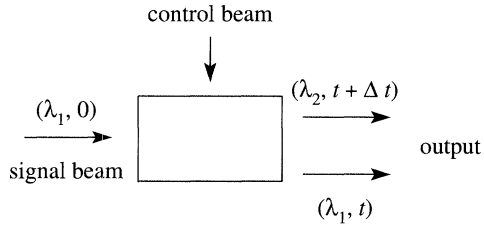

Figure 1. All-optical manipulation of an input signal beam by a control beam resulting in a change of spatial direction (output arrows displaced), of wavelength ($\lambda_1 \rightarrow \lambda_2$), or of time ($t \rightarrow t + \Delta t$).

important feature of any useful device is its throughput, and we define a useful length as α^{-1}. Here we include multiple contributions to the net absorption coefficient α, which is defined as $\alpha = \alpha_1 + \alpha_2 I + \alpha_3 I^2 + \cdots$, where α_1, α_2 and α_3 are the one-photon (linear), two-photon and three-photon absorption coefficients, respectively. Assuming that each in turn can be the dominant cause for loss, the following FOM have been defined:

$$W = \frac{\Delta n}{\alpha_1 \lambda_{\text{vac}}}, \quad T^{-1} = \frac{n_2}{\lambda_{\text{vac}} \alpha_2}, \quad V^{-1} = \frac{n_2}{\lambda_{\text{vac}} \alpha_3 I_c}, \tag{2.1}$$

where I_c is the intensity required for some particular switching operation. For devices that require a nominal 2π phase shift for successful operation, the following inequalities must be satisfied: $W > 1$, $T^{-1} > 1$ and $V^{-1} > 1$. The W FOM can be extended to saturating nonlinearities, i.e. to media in which $\Delta n(I) \rightarrow \Delta n_{\text{sat}}$ as $I \rightarrow \infty$ by replacing Δn by Δn_{sat} and noting that, in the presence of saturation, the required W is essentially doubled. The first FOM is important for both instantaneous and integrating nonlinearities, whereas the remaining two are important for Kerr nonlinearities.

Lossy materials need to satisfy these conditions to be useful for efficient devices. It is possible to have gain in materials, in which case limitations on device performance need to be re-examined. Such an analysis still needs to be performed in detail. There is no *a priori* reason to believe that device performance will be improved, just that there will be no loss.

3. Nonlinear materials

Many nonlinear materials have been discovered (or invented) and investigated since the early days of nonlinear optics. Nevertheless, it is possible to evaluate the FOM discussed above for only a few materials, especially the spectral dependence of these properties. Recent experience has shown that many materials should have spectral windows in which the FOM can be satisfied. This spectral dependence has only been measured for semiconductors, a few polymers and some glasses.

(*a*) *Glasses*

Glasses are amorphous, solid media in which there is no long-range order. The most common example is SiO_2, better known as fused silica. Other glass compositions are known to give higher nonlinearities (Yumoto *et al.* 1993; Newhouse *et al.* 1990; Hall *et al.* 1989). In general, in glasses, all of the electronic transitions are both one- and two-photon active. Therefore, in order to avoid two-photon absorption, it is important to operate at wavelengths $\lambda > 2\lambda_{\text{res}}$ where λ_{res} is the peak of the lowest

Table 1. *Figures of merit of various materials*

(Intensity (assumed) = 1 GW cm^{-2} → 100 W power. $W = n_2 I/\alpha_1 \lambda$ (goal: $W > 1$). $T = 2\lambda\alpha_2/n_2$ (goal: $T < 1$).)

material	n_2 /(cm^2 W^{-1})	α /cm^{-1}	W	λ /T	μm
semiconductors					
GaInAs (c.h.) ($\tau \sim 600$ fs)	4.5×10^{-12}	30–50	0.75	3	1.5
GaInAs (Kerr) ($\tau < 40$ fs)	-3×10^{-12}	30–50	0.5	4	1.5
AlGaAs (0.75 μm)	2×10^{-13}	0.1	8	< 0.3	1.56
organics					
PTS (crystal)	2.2×10^{-12}	< 0.8	> 10	< 0.1	1.60
DANS (polymer)	8×10^{-14}	< 0.2	> 5.0	≈ 0.2	1.32
glass					
SiO_2	2×10^{-16}	10^{-6}	$> 10^3$	$\ll 1$	> 1.06
RN (Corning)	1.3×10^{-14}	0.01	13	< 0.1	1.06
$As_{0.38}S_{0.62}$	4.2×10^{-14}	0.002	16	< 2	1.32

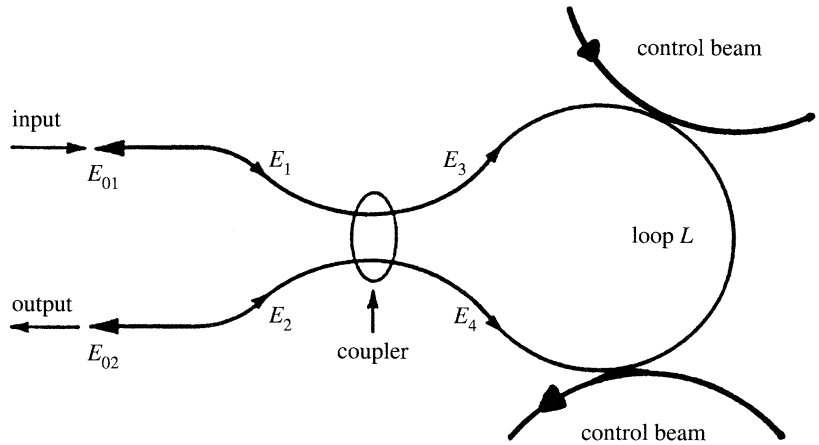

Figure 2. A nonlinear loop mirror in which the output (E_{O2}) is affected by the intensity and relative powers of the counter-propagating beams E_3 and E_4 (via self-phase modulation), or by the power of a control beam (via cross-phase modulation).

energy absorption feature in the linear absorption spectrum. For fused silica, λ_{res} is in the UV so that the FOM are satisfied by fused silica from 0.6 μm right out to a few microns (where linear absorption due to vibrational overtones comes into play via the W FOM). On the other hand, chalcogenide glasses, which have a much higher nonlinearity than fused silica, have a linear absorption cut-off wavelength of about 0.6 to 0.65 μm, depending on the composition (Asobe *et al.* 1993). In fact, this is a recurring theme. The longer the λ_{res}, the larger the resonant and non-resonant nonlinearity. These nonlinearities are non-resonant and respond on fs time scales.

Values of the FOM are listed in table 1 for a few glasses. Although the nonlinearity

n_2 has been measured for a much larger number of glasses than listed here, the other parameters needed to evaluate the FOM are not known. The trade-off between nonlinearity and FOM is clear. The higher the nonlinearity, the worse the FOM. Nevertheless, many glasses do have properties which are very attractive for all-optical devices, if they can be fabricated into low loss fibres.

Silica fibres have been used extensively for signal processing applications using data in pulse-coded format (Agarwal 1989). Although many different geometries have been used for switching, demultiplexing, logic, etc., the most common appears to be the nonlinear loop mirror (NOLM), sketched in figure 2. Operation relies on a different nonlinear phase shift being imparted on the two oppositely propagating beams so that the interference condition at the output junction can be changed by changing the optical power of either a control beam (typically injected and removed within the loop), or the inputs into the loop. By choosing fibres with appropriate group-velocity dispersion, at either 1300 or 1550 nm, signal processing operations can be performed using temporal solitons. Not only does this make the device operation compatible with soliton transmission lines, it also eliminates distortion in the envelope of the output pulse which can occur in non-solitonic systems.

(*b*) *Semiconductors*

Semiconductors have been investigated extensively for all-optical processing applications, dating back to the early days of bistability. Essentially, the four following mechanisms for producing the required nonlinearity have been used.

(i) *Band-filling and exciton bleaching for photon energies below the bandgap*

The absorption of one or more photons leads to a change in the carrier density in the valence and conduction bands and, hence, in the refractive index. Here the trade-off between index change and absorption inherent in the W and T parameters, as well as the carrier relaxation (or sweep out) times are important (for a review see Stegeman & Miller 1993). The FOM trade-offs are summarized in figure 3. It is clear that detuning from the bandgap leads to the desired increasing values of W. However, for too large a detuning the T parameter becomes important. The recovery time of the nonlinearity can be reduced to tens of ps by applying a strong electric field to sweep the generated carriers out of the optical path.

A number of devices have been implemented based on these nonlinearities at wavelengths just above the bandgap (for a review see Stegeman & Miller 1993). Because of the flexibility afforded by its two input and two output ports, the most common device has been the nonlinear directional coupler (NLDC), illustrated in figure 4. For example, this geometry can be used for all-optical switching, demultiplexing and multiplexing of data streams, optical logic, etc., and indeed some of these have been demonstrated in this material system.

(ii) *Non-resonant nonlinearity for photon energies below one half of the bandgap*

Here the important trade-offs are found in the T and V parameters (for a review see Stegeman *et al.* 1994). The principal interest has been in the 1550 nm communications window with $Al_xGa_{1-x}As$, where $x = 0.18$ gives a half bandgap photon energy at 1500 nm. The T–V ($\Delta\phi^{NL} = 2\pi$ assumed for V) trade-off is shown in figure 3. There is clearly a photon energy window just below half the bandgap where this material behaves like a classic Kerr nonlinearity. Its response time is less than 100 fs.

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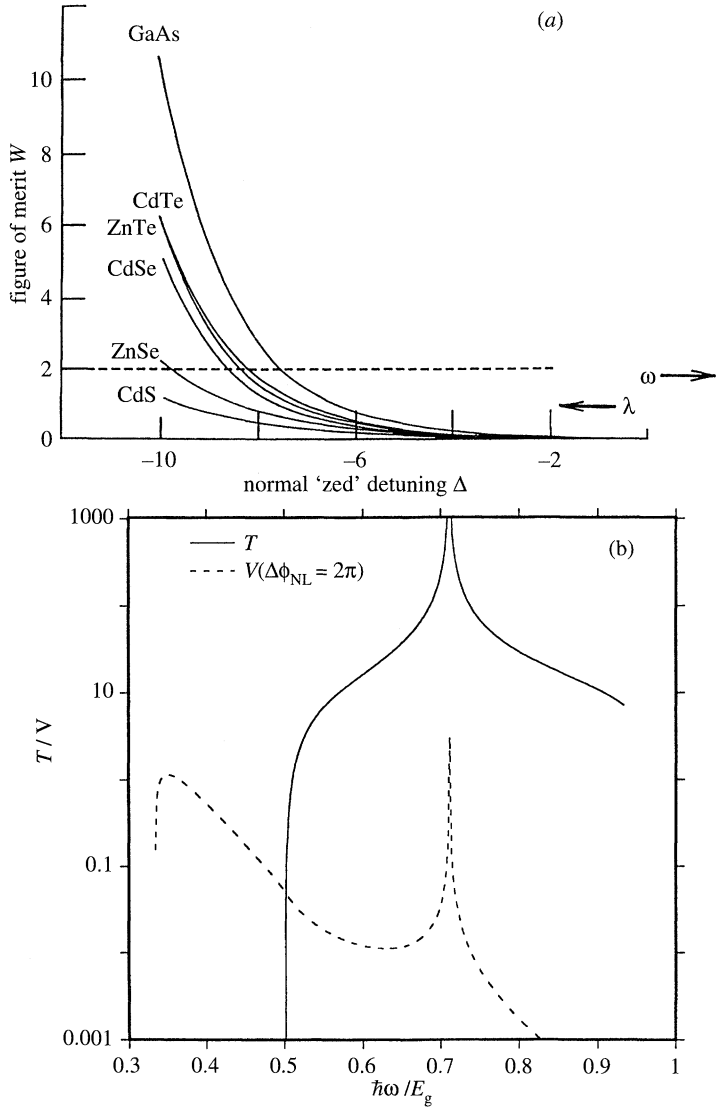

Figure 3. (*a*) FOM W ($= \Delta n_{\mathrm{sat}}/\alpha_1\lambda$). (*b*) FOMs T ($= \alpha_2\lambda/n2$) and V ($= \alpha_3 I_c\lambda/n_2$), where I_c is a switching power corresponding to $\Delta\phi^{\mathrm{NL}} = 2\pi$) for semiconductors. Δ is the detuning from the bandgap, normalized to the exciton linewidth. E_g is the bandgap energy and $\hbar\omega$ is the photon energy.

A large number of all-optical devices have been demonstrated in this material system, including NLDCs, X-junctions, Mach–Zehnder interferometric switches, zero gap couplers etc. (for a review see Stegeman *et al.* 1994). Of these, the NLDCs have proven the most powerful, as discussed above. Efficient and distortion-free multiplexing and demultiplexing of data pulses has been demonstrated.

(iii) *Ultrafast refractive nonlinearity near transparency in active semiconductor amplifiers*

The term transparency refers to an electrical pumping condition in which the gain and absorption are just balanced for the light-carrier interaction. Multiple nonlin-

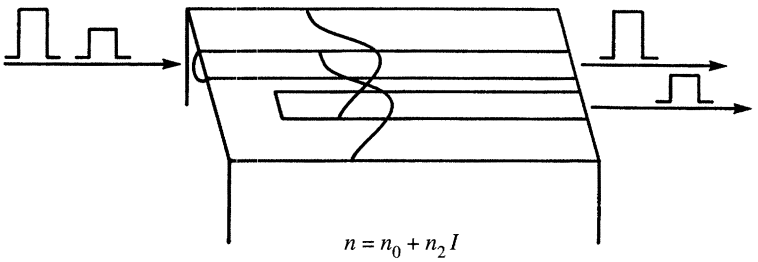

Figure 4. Schematic of a nonlinear guided wave directional coupler. Shown is self-switching, in which the output is determined by the intensity of the input pulse.

earities occur, including the ultrafast Kerr nonlinearity ($n_2 \sim -4 \times 10^{-11}$ cm^2 W^{-1}) which recovers on a < 40 fs time scale, spectral hole burning (~ 100 fs) and carrier heating ($n_2 \sim 5 \times 10^{-12}$ cm^2 W^{-1}, $\tau \sim 600$ fs) (D'Ottavi *et al.* 1994; Hall *et al.* 1993). This approach is quite new and the switching devices investigated were not necessarily optimized for nonlinear FOM. To date, such amplifiers have exhibited large linear and nonlinear loss near transparency so that both W and T appear to be marginal for applications (see table 1). The only device reported to date has been the NLDC, the operation of which was verified, but the results were far from optimal due to large losses (Davies *et al.* al 1993, Lee *et al.* 1993).

(iv) *Carrier nonlinearities in active semiconductor amplifiers*

It is this case which is most likely to be adopted in practical devices, and, in fact, device demonstrations have been very impressive to date (for a review see Adams *et al.* 1995). In the region of gain achieved by pumping the carriers into the conduction band, large nonlinear phase shifts can be obtained for an incident beam when electron transitions occur from the conduction to valence bands. For example, only 3 pJ pulse energies are required for π phase shifts in 0.5 mm long amplifiers. The recovery time is given by either the carrier sweep-out time, or the carrier recombination time. Despite the limited recovery time, active amplifiers have been used in clever ways to perform ultrafast switching operations. For example, demultiplexing was achieved by placing the semiconductor amplifier just off centre in a loop mirror so that the counter-propagating beams interact asymmetrically with a carrier distribution excited by an injected control beam (Eiselt 1992; Sokoloff *et al.* 1993). The key parameters here are T and the relaxation times.

One of the most interesting applications is to wavelength shifting. The basic idea is shown in figure 5 (Manning & Davies 1994). Pumping at λ_1 brings the system gain into saturation. An input at the wavelength λ_2 modulates the gain and therefore an input signal at the wavelength λ_3. The role of the pumping beam is to maintain the semiconductor at saturation so that the recovery time becomes independent of the signal spacing in time, leading to recovery times of tens of ps. This clearly operates as a wavelength shifter, valuable for communications applications.

(*c*) *Polymers*

Organic materials, usually in the form of polymers, can have large third-order nonlinearities. This is a consequence of electron delocalization along molecular chains characterized by double and triple carbon bonds. That is, it is the $2p_z$ electrons associated with the original carbon atomic orbitals that stick out of the plane of the molecules and overlap in space, facilitating electron motion over long effective

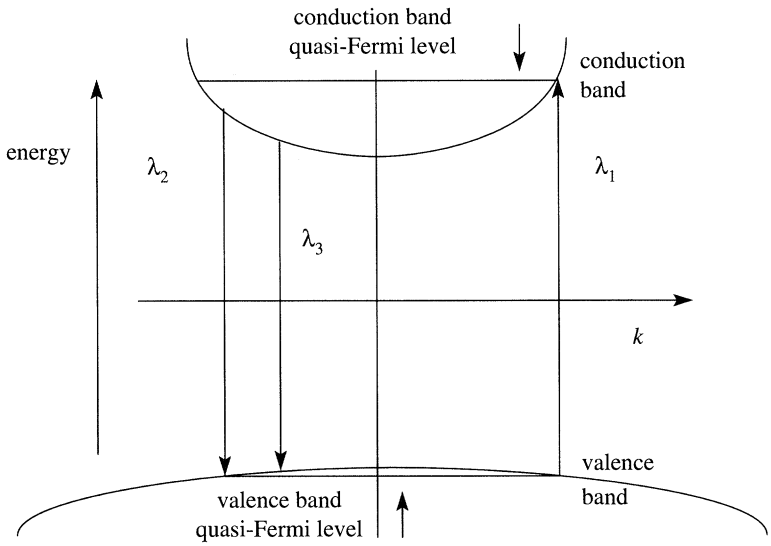

Figure 5. Simplified energy diagram illustrating the principle of wavelength shifting. λ_1 is the holding beam wavelength; λ_2 is the input signal wavelength; and λ_3 is the output wavelength-shifted signal.

distances down the chain. Although this delocalization mechanism was recognized back in the 1970s and the effects of delocalization and electron correlation on the molecular electronic structure have been under investigation since then, it is only in the last few years that parameters such as n_2 and α_2 have been measured at different wavelengths (Lawrence *et al.* 1994*a*, *b*).

Although many different molecular structures have been investigated by techniques such as third-harmonic generation, degenerate four-wave mixing, etc., only two classes of materials are sufficiently well studied to evaluate FOMs. The first are symmetric molecules which lack a permanent dipole moment; for example, linear molecules. The electronic states are either one- or two-photon allowed, and electromagnetic coupling to both types is important in defining the nonlinearity. For example, in the single-crystal polymer paratoluene sulfonate (PTS), the nonlinearity n_2 ($\propto \mathrm{Re}[\chi^{(3)}(-\omega;\omega,-\omega,\omega)]$) and effective two-photon coefficient α_2 ($\propto \mathrm{Im}[\chi^{(3)}(-\omega;\omega,-\omega,\omega)]$) have been measured on the long wavelength side of the dominant one-photon resonance. This has allowed the T FOM to be evaluated for wavelengths between 1000 and 1600 nm, overlapping the two communications windows at 1300 and 1550 nm (Lawrence *et al.* 1994*a*, *b*). From figure 6 it is clear that $T < 1$ in both windows. Furthermore, the non-resonant nonlinearity far from any absorption line is one of the largest known (2.2×10^{-12} cm^2 W^{-1}). So why was this material not adopted immediately for signal processing? The problem is the scattering due to defects etc., which limits the available propagation distance. A major effort is needed to improve the material processing to obtain an optical quality material suitable for applications. Another material which has exhibited large n_2 and manageable α_2 (i.e. $T < 1$) is PPV around 800 nm (Luther-Davies *et al.* 1995). Again, however, material processing to produce a low scattering loss material is currently the major barrier.

There have also been sporadic measurements on charge-transfer molecules in polymer hosts which were developed originally for electro-optic applications. For example,

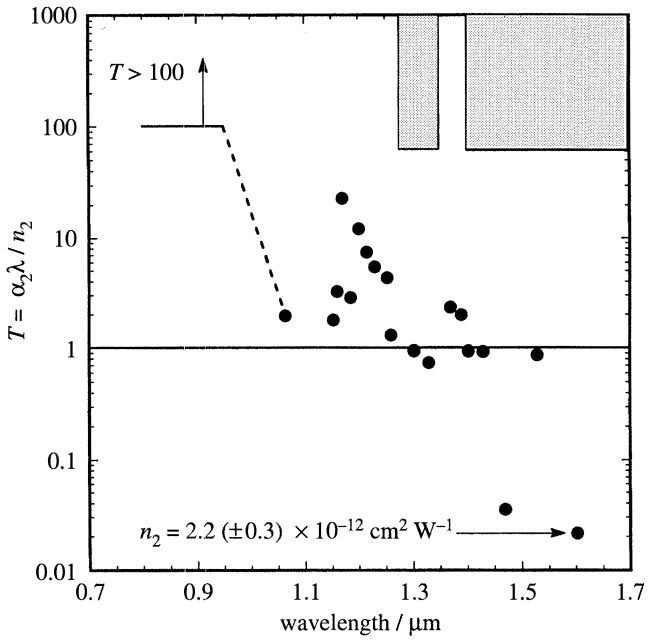

Figure 6. The two photon FOM T versus wavelength for the single-crystal polymer PTS. The non-resonant value of the nonlinearity n_2 is also given.

at 1300 nm, DANS exhibits $T < 1$ and $W > 1$ (Kim *et al.* 1993). However, this class of molecules exhibits nonlinearities n_2 in the range $(2 \rightarrow 0.5) \times 10^{-13}$ cm^2 W^{-1}, down by an order of magnitude from the previously discussed PTS case and comparable with the well-established technologically compatible AlGaAs.

Certainly, further research is needed in this promising class of materials.

4. 'Cascading'

As discussed previously, a nonlinear phase shift is the key requirement for most all-optical devices. It is possible to obtain such phase shifts from second-order nonlinear interactions. That third-order effects can be mimicked by successive second-order processes has been known since the early days of nonlinear optics. However, it is only recently that the potential of this approach has been investigated, driven to a large degree by the development of new materials (primarily organic) with large second-order nonlinearities, and by new ways of utilizing the large diagonal nonlinearities for phase-matching in existing second-order materials.

The concept of 'cascading' is summarized in figure 7. In a second-order material ($d^{(2)}(-2\omega;\omega,\omega) \neq 0$), both up-conversion ($\omega + \omega \rightarrow 2\omega$), better known as second harmonic generation (SHG), and down-conversion ($2\omega - \omega \rightarrow \omega$) occur. The maximum SHG efficiency depends inversely on the phase velocity mismatch $(v_2 - v_1)^{-1}$ between the fundamental and second-harmonic waves. That is, the phases of the second-harmonic and fundamental beams evolve differently in space. Therefore, when the second-harmonic photon is down-converted back to the fundamental after some propagation distance, typically the coherence length L_c, the net phase of the fundamental beam is changed, i.e. there is a $\Delta\phi^{NL}$. The phase shift is nonlinear because the stronger the conversion to SHG (which depends on the input fundamental inten-

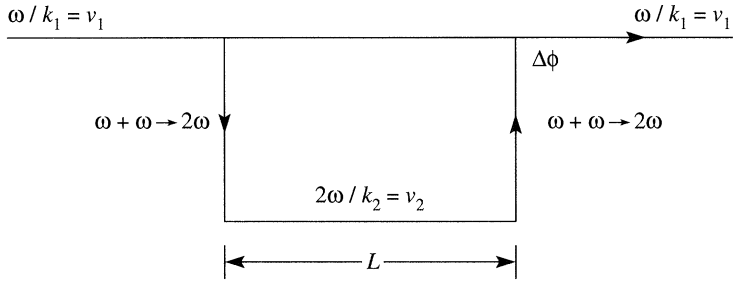

Figure 7. Schematic representation of cascading. Second-harmonic radiation is generated, propagates a distance L ($\equiv$ coherence length), and converts back to the fundamental beam. In the wavevector mismatched case, the fundamental and harmonic phase fronts have evolved differently so that the returning fundamental is out of phase with the non-converted fundamental, shifting its net phase.

Table 2. *Effective optimized nonlinear coefficients n_2 via cascading for materials with representative d_{ij}*

material $L = 1$ cm	d_{ii} /(pm V^{-1})	d_{ij} /(pm V^{-1})	n_2 (effective) (cm^2 W^{-1})
$LiNbO_3$	36		2×10^{-11}
$LiNbO_3$		5.8	5×10^{-13}
MNA	165		7×10^{-10}
NPP		84	2×10^{-10}
DAST	600		6×10^{-9}

NPP: N-(4-Nitrophenyl)-(L)-prolinol.
MNA: 2-Methyl-4-nitroaniline.
DAST: dimethyl amino stilbazolium tosylate.

sity), the larger the net phase shift. We note, however, that the phase shift depends on the fundamental input intensity only in the limit of negligible SHG conversion: for large SHG conversion the phase shift becomes linear in the input field (Stegeman *et al.* 1993). Finally, it is clear that $\Delta\phi^{\mathrm{NL}}$ can be either positive or negative, depending on which of v_2 or v_1 is larger.

The principal reason why this cascading mechanism has become 'fashionable' is summarized in table 2. In the limit of weak SHG, it is possible to define an effective n_2 to quantify cascading, i.e.

$$n_{2,\mathrm{eff}} \simeq \mathrm{sign}(\Delta v)\frac{4}{\epsilon_0 c}\frac{[d_{\mathrm{eff}}^{(2)}]^2}{n^3}\frac{L_{\mathrm{c}}}{\lambda}, \tag{4.1}$$

and the values listed for a number of materials appear very promising. However, it must be emphasized that proximity to wavevector matching is a necessity and this has not been achieved yet in many of the most promising materials listed in this table.

A number of experiments have been reported already verifying the details of the cascaded nonlinearity. For example, a direct measurement of a large nonlinear phase shift was obtained near type-I phase-matching in a $LiNbO_3$ channel waveguide. The key point is that a nonlinear phase shift of 1.5π was obtained. One of the potential

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criticisms of this approach to generating nonlinear phase shifts is that power is lost from the fundamental beam to the second-harmonic beam, and this of course must happen because an exchange of photons is necessary. However, if the wavevector mismatch is not uniform along the propagation path, then the SHG conversion efficiency and phase shift can be controlled independently (Schiek *et al.* 1994).

This area is clearly in its infancy and still needs to be critically assessed. It is noteworthy, however, that an all-optical Mach–Zehnder switching device has already been achieved.

5. Summary

There are now a number of materials options for using nonlinear optics for signal processing. The most commonly used are silica fibres. Although their nonlinearities are weak, the very low propagation losses allow km lengths to be used. As long as the delay associated with the transit time is not a problem, peak switching powers of mW have been demonstrated. Coupled with the availability of erbium-doped fibre amplifiers, fibre devices are currently the option of choice.

The application of carrier nonlinearities associated with electrically pumped active semiconductor amplifiers is growing rapidly. The devices have small switching energies (a few pJ) coupled with very short device lengths (less than a mm). Now that recovery times of tens of ps are feasible with low power cw control beams; these devices are capable of operation at tens of Gbits s^{-1}. The ability to also efficiently shift the signal wavelength is very useful and these devices will at least partially replace devices based on fibre nonlinearities.

To date, other semiconductor approaches have not proven to have the right combination of parameters for practical devices. For example, using the nonlinearities below one half of the bandgap results in classical Kerr law devices. However, the switching energies (referred to 1 ps pulses) are just too high, and, although this material system is and will continue to be a wonderful laboratory for investigating new nonlinear optical phenomena, it is unlikely to be used for practical devices.

There currently appear to be limitations to using ultrafast nonlinearities in active semiconductor amplifiers. Both the linear and nonlinear losses are currently issues, and the switching energies, again referenced to 1 ps pulses, appear to be many tens of pJ. However, many improvements appear to be possible here and it will be interesting to see where this approach finally ends up.

It is just too early to tell whether device applications of organic materials and the cascading phenomenon will be realized. Both approaches offer large nonlinear coefficients. For polymers the problem with scattering has already been identified. In the case of cascading, this approach is so new that the potential complications are still unknown.

This work has been supported by the National Science Foundation, the Air Force Office of Scientific Research and the Advanced Projects Agency through the Army Research Office.

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