# NONLINEAR OPTICS

Self-Reflection and Routing of Multicolor Solitons

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The reflection and transmission of optical waves incident onto a boundary between two linear materials are governed by the well-known Snell's law and Fresnel equations. In optically nonlinear media, however, since the intensity of the light itself changes the optical properties of the boundary, the classical equations are no longer valid.[1] This yields a fascinating variety of new phenomena, which include the power-dependent reflection of light beams at the interface between different cubic nonlinear media. Since quadratic nonlinearities also cause self-effects on light beams, similar phenomena were predicted at the interface between two quadratic materials with different properties. A dramatic example is encountered when only the nonlinearity varies at the interface, a situation that can also change the reflection and transmission properties even when the refractive index is continuous across the boundary.[2] Such is the case, e.g., at the interface in a ferroelectric medium that separates two periodically poled regions with poling patterns shifted by, e.g., a half of a period relative to each other [Fig. 1(a)]. Whether an incoming multi-color soliton beam is reflected from or transmitted through such an interface was predicted to depend on the magnitude of the input light intensity and on the angle of incidence at which the soliton hits the interface. At low intensities and/or large incident angles, the soliton should pass through the boundary, whereas at high intensities and at small incident angles it was predicted that the soliton would self-reflect.[2] This type of beam reflection is unique to solitons since only nonlinear waves are affected by the induced potential well, and since it is their particle-like nature that allows solitons to retain their entity after reflection. As part of a multiteam effort, we have
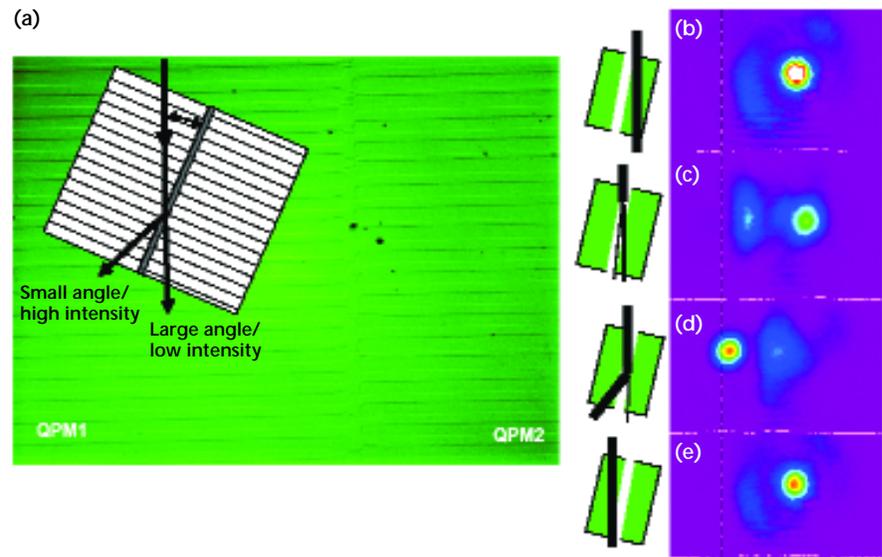
Figure 1. Microscope picture of the sample quasi-phase-matched gratings and the boundary (*inset: geometry of the light beam-sample interaction*) and collage of beam geometries (*right-hand side*) and output beam distributions for different relative sample-input beam positions. Input intensity 6 GW/cm^2 and incident angle of approximately 0.5 degrees.

been able to design and fabricate a suitable sample to test, for the first time to our knowledge, this kind of phenomena and thus to verify the existence of the intensity dependent nonlinear reflection.[3]

The experiments were conducted under conditions of phase-matched second-harmonic generation pumped with 25-ps laser pulses in a periodically poled, 7.5-mm-long potassium titanyl phosphate crystal that corresponds to five diffraction lengths inside the crystal. A nominal 9-μm poling period gave noncritical phase matching at 1064 nm at room temperature. The first step was to verify the formation of spatial solitons in this material and to characterize their features.[4]

Solitons were formed with input beams with a waist of approximately 16 μm with input intensities ≈ 5 GW/cm^2. Then we tested the soliton reflection at the nonlinear interface. The key experimental observation is illustrated in Fig. 1(b-e), which shows the output beams that we obtained by translating the dislocated sample from left to right. That the soliton is reflected back after it

hits the nonlinear interface is clearly visible in Fig. 1(d). We refer the reader to Ref. 3 for complete details.

This observation, which culminates a multiyear theoretical effort by several groups around the world addressed at different strategies aimed at soliton control by quasi-phase-matching engineering,[5] opens the door to the implementation of a number of soliton processing schemes. These include the power-controlled steering and trapping of solitons by use of a variety of different potential wells. The limit is set only by the structures that can be imagined and created by the powerful technique of quasi-phase-matching engineering.

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Supraoptical Nonlinearity of Liquid Crystals

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Researchers studying nonlinear optical materials have made tremendous strides in recent years, achieving new spectral and temporal power and geometric size frontiers on an almost daily basis. The nonlinear optical response of nematic liquid crystals (NLCs), self-organizing molecular systems that exhibit optically induced collective crystalline axis reorientation, is a noteworthy example. Recent studies[1] have shown that their extremely strong light scattering properties can be used for self-action effects with unprecedented low optical power in very thin film. Specifically, an all-optical polarization switching effect has been demonstrated with a 1.55-μm laser as well as with visible lasers (see Fig. 1), in which a milliwatt power incident beam is almost completely converted to an orthogonally polarized output after it traverses a 200-μm-thick NLC film. With an optimal choice of materials, the operating power could easily be reduced to a sub-milliwatt level. Liquid crystals have also ushered in the era of supranonlinearities characterized by an optical index change coefficient $n_2 \gg 1$ cm^2/W ($n_2 = \Delta n/I$, where Δn is the index change and I is the optical intensity). Such nonlinearities have recently been discovered in Methyl Red dye molecules, C60 and carbon nanotube doped NLC.[2] These supra-optical nonlinearities have enabled many nonlinear processes, e.g., optical limiting and image processing,[3] with even lower operating power thresholds, which makes these NLC films competitive alternatives to their much more costly counterparts, optoelectronic devices.

What then, it comes naturally to ask, is the limit on such supranonlinearity? A glimpse into the answer could be gained by considering the basic light-LC interaction[4] as depicted in Fig. 1. The energy density involved in reorienting the LC axis by angle θ is

$$U \left[\frac{\text{erg}}{\text{cm}^2} \right] \approx K \left(\frac{\partial \theta}{\partial x} \right)^2 L,$$

Figure 1. Schematic depiction of a laser interacting with a nematic liquid crystal film. Scattering by the director axis fluctuation gives rise to a spectrum of new spatial and temporal frequency components. A "Stoke-shifted" noise component experiences maximum gain and grows to a coherent beam.

where L is the interaction length and K is the LC elastic constant. In a grating diffraction experiment, θ is of the form $\theta = \theta_0 \sin qx$, where $q = 2\pi/\Lambda$. Thus $U \sim K \pi^2 \theta^2 L/\Lambda^2$. On the other hand, the energy provided by the light beam is $E = I\tau [1 - \exp(-\alpha L)] \sim \alpha LI\tau$, where α is the absorption coefficient and τ is the response time. Equating E and U, i.e., assuming complete conversion of absorbed light energy to reorientation, we obtain $I\tau \approx K \pi^2 \theta^2/\alpha\Lambda^2$. For an interaction geometry as depicted in Fig. 1, the change in index experienced by an incident extraordinary wave is

$$\Delta n \sim (n_e - n_o)\theta^2 \sim (n_e - n_o) \ I\tau \ \alpha\Lambda^2/K.$$

Writing $\Delta n = n_2 I$ yields $n_2 \sim (n_e - n_o)\tau \ \alpha\Lambda^2/K \pi^2$. The value of n_2 and the response time can vary significantly depending on parameters such as birefringence and viscosity, sample thickness and other factors such as laser intensity, the presence of other applied fields and the actual process involved. In NLCs, τ is typically of the order of tens of milliseconds (10^{-2} s) for $\Lambda \sim 20$ μm. Using $K \sim 10^{-7}$ erg/cm, $(n_e - n_o) \sim 0.2$, and $\alpha \sim 100$ cm^{-1}, yields $n_2 \sim 1$ cm^2/W. Even larger n_2 values can be expected. For example, if $\Lambda \sim 100$ μm, $\tau \sim 1$ s, $K \sim 10^{-7}$, $(n_e - n_o) \sim 0.4$ and $\alpha \sim 200$ cm^{-1}, we obtain $n_2 \sim 1,000$ cm^2/W.

In the near future, we can expect other favorable surprises and nonlinear optical wonders to emerge from studies of liquid crystals.

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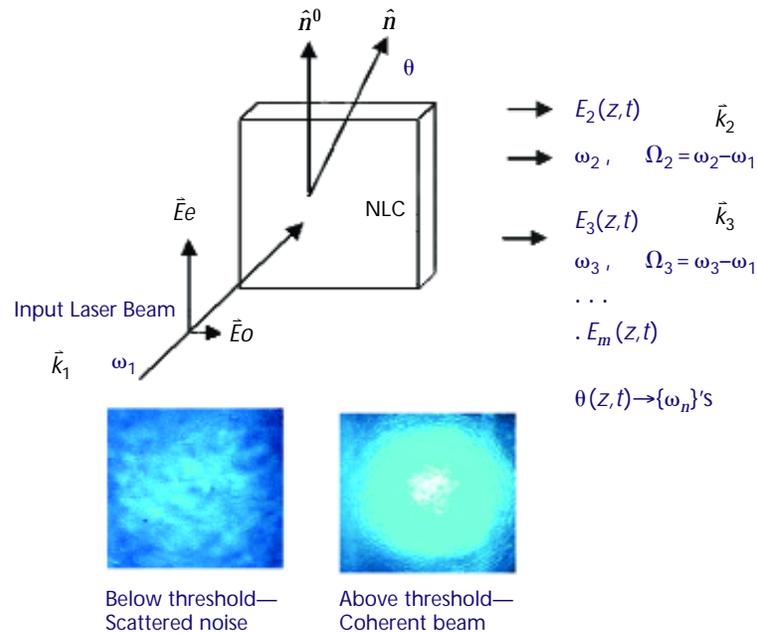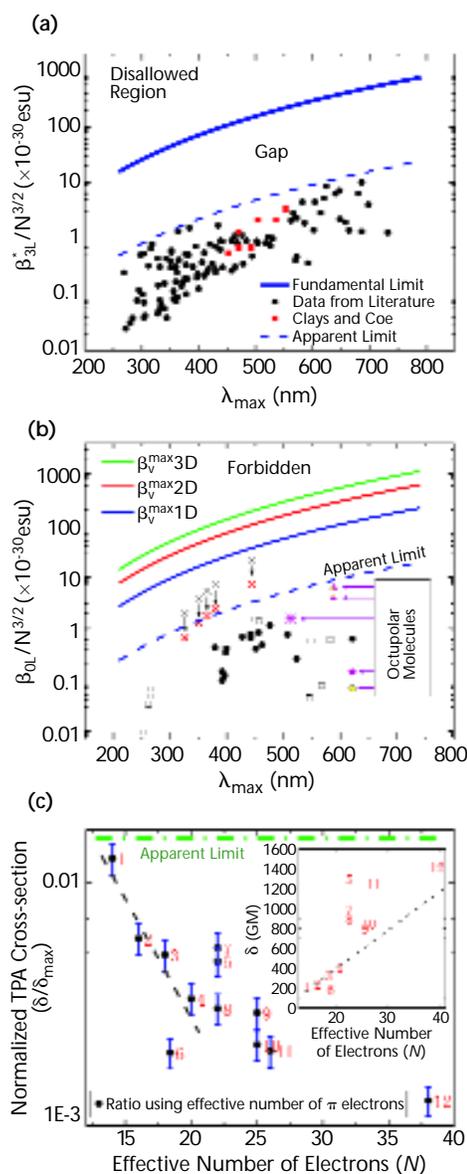
(a)

(b)

(c)

Figure 1. (a) Normalized β values from the literature (*points*), maximum limit (*solid line*) and apparent limit (*dashed line*). (b) Normalized β values from the literature (*points*) and calculated limits of β (*curves*) for 1D, 2D and 3D molecules. (c) TPA cross section (*inset*) and cross section divided by its maximum limit (*points in main graph*).

Fundamental Limits of Nonlinear Susceptibilities

Mark G. Kuzyk

The nonlinear susceptibility $\chi^{(n)}$ is a property of matter that describes the interaction of matter with light. For example, $\chi^{(2)}$ governs such diverse phenomena as second harmonic generation and electro-optic switching, while $\chi^{(3)}$ is responsible for all-optical switching, optical limiting and self-focusing mode-locking in a Ti:sapphire laser. Thus, the answer to the question, "are there fundamental limits to the nonlinear susceptibilities," has profound implications for both the potential characteristics of future devices, and basic science. Our research answers the question.

Nonlinear susceptibilities can be calculated by use of quantum perturbation theory, which leads to a complicated expression often called the sum-over-states (SOS) equation. Typically, one calculates the hyperpolarizabilities β, γ, …, which are the microscopic analogs of $\chi^{(2)}$, $\chi^{(3)}$, … (There are simple models that relate microscopic values to bulk values.) Since the SOS expression includes infinite sums over the quantum states of a system, there was previously little hope of unraveling the mysteries of what makes a large nonlinear susceptibility. In our work, we used the Thomas Kuhn sum rules—an infinite set of equations, each having an infinite number of terms—to collapse the SOS expression into a surprisingly simple form. For the first hyperpolarizability β, we obtain

$$\beta_{\max}^{(Gauss)} = 559 \times 10^{-30} N^{3/2} \cdot \lambda^{7/2} \, (\mu m),$$

where β is in Gaussian units, N is the number of electrons in the system and λ is the wavelength of maximum absorption of the first excited state in micrometers. A similar simple equation was found for other susceptibilities.[4]

Figure 1(a) shows a plot of β for those organic molecules with the largest susceptibilities as tabulated from the literature.[3] All the points fall below the maximum fundamental limit. There also appears to be another limit (dashed line), which is a factor of $10^{-3/2}$ lower than the fundamental limit. Figure 1(b) shows a plot that includes octupolar

molecules, which were thought to have larger β than dipolar ones.[2] The lines show calculated limits for one-dimensional (1D), two-dimensional (2D) and three-dimensional (3D) molecules. All the measurements fall comfortably below the 1D limits, and most points fall below the apparent limit except for the crosses ($\times$), which were errant measurements that were later corrected (vertical arrows). Figure 1(c) shows two-photon absorption (TPA) cross sections.[5] Although the absolute value of δ increases as more electrons are added to the system (inset), the normalized TPA cross section $\delta/\delta_{\max}$ decreases (main plot). Contrary to the present paradigm, the smaller molecules appear to use their electrons more effectively. The TPA cross section also seems to be subject to an apparent limit of $10^{-3/2}$ times the fundamental limit.

These calculations could lead to a rethinking of the paradigms now used to design molecules. The theoretical results are useful to the device engineer as a guide for understanding the best material properties that can be achieved, to the chemist as a metric of the usefulness of a particular synthetic paradigm and to the physicist for an understanding of how complex material structures could be used to obtain as much out of a material as is fundamentally possible.

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