Electrostrictive contribution to the intensity-dependent refractive index of optical fibers

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We show that electrostriction contributes significantly to self-action effects in optical fibers, adding 19% to the nonlinear refractive index for fields that vary slowly compared with the ~1-ns time scale of the acoustic response. Electrostriction also modifies the tensor nature of the nonlinear-optical response. The electrostrictive nonlinearity is the origin of the observed difference between measurements of n_2 with cw and mode-locked lasers. © 1996 Optical Society of America

The nonlinear refractive index of optical fibers continues to draw considerable attention. With the prospect of densely packed wavelength-division-multiplexed channels and soliton transmission,[1] the ability to predict and ultimately to control nonlinear interactions in fibers becomes increasingly important. The first measurement of n_2 in optical fibers utilized self-phase modulation of 100-ps pulses.[2] Subsequently, various measurement techniques relying on self-phase modulation (SPM) or cross-phase modulation (XPM) with either mode-locked pulses or slowly modulated cw signals have been reported,[3–6] with increasing emphasis on accuracy and precision.

Results of these measurements have typically been analyzed with the assumption that the origin of the nonlinearity was nonresonant electronic response. This assumption leads to specific predictions regarding polarization properties and specifically precludes the possibility of time-scale effects. However, calculations predict that the real part of the Raman susceptibility will contribute 15% of the total nonlinearity measured in the SPM experiments,[7] and this contribution is essentially instantaneous on the time scale of the pulses. Electrostriction, the process in which the material density increases in response to the intensity of an applied optical field, has a much slower response time, approximately the 1-ns transit time of a sound wave propagating across a fiber core. The influence of electrostriction was previously analyzed for long-range soliton interactions.[8] Self-action effects owing to electrostriction have generally been assumed to be negligible, but we show below that electrostriction can add measurably to the nonlinear response.

In the perturbation approximation the refractive-index change associated with electrostriction is given by $\Delta\tilde{n}(t) = \Delta\tilde{\epsilon}(t)/2n$, where $\Delta\tilde{\epsilon}(t) = (\partial\epsilon/\partial\rho)\Delta\tilde{\rho}(t)$ is the change in the dielectric constant $\Delta\tilde{\epsilon}(t)$ that is due to the change in density $\Delta\tilde{\rho}(t)$ and n is the linear refractive index. To determine the refractive-index change we first solve the acoustic wave equation to determine the impulse response function $h(t)$ that describes the density variation initiated by a delta-function intensity pulse. The general solution for the density variation is the convolution of $h(t)$ with the intensity of the applied field, and thus the refractive-index change has the form

$$\begin{aligned}\Delta\tilde{n}(t) &= (\partial\epsilon/\partial\rho)\Delta\tilde{\rho}(t)/2n \\ &= \frac{1}{2n}(\partial\epsilon/\partial\rho)\int_0^t \mathrm{d}t' h(t-t')\,|\tilde{\mathbf{E}}(t')|^2. \qquad (1)\end{aligned}$$

The acoustic wave equation is given by[9]

$$\frac{\partial^2\Delta\tilde{\rho}}{\partial t^2} - \Gamma'\nabla^2\frac{\partial\Delta\tilde{\rho}}{\partial t} - v^2\nabla^2\Delta\tilde{\rho} = -\frac{\gamma_e}{4\pi}\nabla_\perp^2|\tilde{\mathbf{E}}|^2. \qquad (2)$$

Here v is the sound velocity in the fiber, Γ' is the acoustic damping parameter, $\gamma_e = \rho_0(\partial\epsilon/\partial\rho)$ is the electrostrictive coefficient, and we use c as the vacuum speed of light. We solve Eq. (1) in the Fourier domain[8] for $\Delta\rho(\Omega, q)$ as a function of the hypersonic frequency Ω and the transverse wave vector q. The frequency-response function $H(\Omega)$ is obtained from $\Delta\rho(\Omega, q)$ after integration over q. From the convolution theorem, the frequency- and time-dependent index changes associated with the electrostrictive response to an arbitrary pulse with intensity spectrum $B(\Omega)$ are thus found to be the Fourier-transform pair

$$\Delta n(\Omega) = n_2(\mathrm{str})I_0 B(\Omega)H(\Omega),$$

$$n_2(\mathrm{str}) = \frac{1}{8}\frac{1}{c\rho_0}\left(\frac{\gamma_e}{nv}\right)^2, \qquad (3a)$$

$$\Delta\tilde{n}(t) = n_2(\mathrm{str})I_0\int_{-\infty}^{+\infty} B(\Omega)H(\Omega)\exp(-i\Omega t)\mathrm{d}(\Omega/2\pi). \qquad (3b)$$

We have defined the field such that $\tilde{\mathbf{E}} = \mathbf{E}_0 b(t) \times \exp(-i\omega_0 t) +$ c.c., with a peak optical intensity given by $I_0 = (nc/2\pi)|\mathbf{E}_0|^2$. The relevant spectrum is $B(\Omega) = \int_{-\infty}^{\infty}|b(t)|^2\exp(i\Omega t)\mathrm{d}t$, where the envelope function $b(t)$ has a maximum value of unity. Typical parameter values for optical fibers are $n = 1.46$ for the linear refractive index, $\epsilon = 2.1$ for the dielectric constant, $\rho_0 = 2.2 \times 10^{-3}\ \mathrm{kg/cm^3}$ for the density, and $v = 5.9 \times 10^5\ \mathrm{cm/s}$ for the speed of sound. The electrostrictive coefficient can be estimated as $\gamma_e = (\epsilon - 1)(\epsilon + 2)/3 = 1.5$, which follows from the Lorentz–Lorenz law for the dependence of the dielectric constant on material density. We calculate that $n_2(\mathrm{str}) = 0.574 \times 10^{-16}\ \mathrm{cm^2/W}$.

Although we do not have an analytic expression for the frequency-response function $H(\Omega)$, we can

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facilitate the numerical integration of $\Delta\rho(\Omega, q)$ over q by introducing a normalized sound velocity $\nu = v/\Gamma a$, acoustic wave vector $x = qa$, and acoustic frequency $g = \Omega/\Gamma$, where the phonon damping rate is $\Gamma = 3 \times 10^{-7}\ \mathrm{s}^{-1}$. We assume azimuthal symmetry in the density function and treat the cladding as infinite, and we ignore the longitudinal components of the Laplacian, as the transverse gradient of the intensity dominates for all but subpicosecond pulses. $H(\Omega)$ can then be expressed as the dimensionless integral

$$H(\Omega) = \nu^2 \int_0^\infty \frac{x^3 \exp(-x^2/2)}{(\nu^2 x^2 - g^2) + ig^2}\, \mathrm{d}x\,, \tag{4}$$

which is normalized such that $H(0) = 1$.

From Eqs. (3) it is evident that the influence of the electrostrictive nonlinearity depends strongly on the duration and the intensity spectrum of the pump pulse. Figure 1 is a plot of the relative strength of the electrostrictive nonlinearity $\eta = \Delta\tilde{n}(t)^{\max}/[n_2(\mathrm{fast})I_0]$ versus the pulse width for Gaussian and square temporal profile pulses, assuming that $n_2(\mathrm{fast}) = 2.96 \times 10^{-16}\ \mathrm{cm}^2/\mathrm{W}$ for SPM of linearly polarized light.[3] The electrostrictive response to a cw signal is 19% of the fast nonlinearity, or 16% of the total nonlinear response. This result holds for linearly polarized light propagating through a medium in which the polarization state is preserved.

In general, however, the polarization state is not preserved in propagation through optical fibers because of the effects of linear and nonlinear birefringence. It is therefore necessary to determine the effective value of n_2 for propagation through a non-polarization-preserving fiber.[10,11] Furthermore, the tensor properties of electrostriction differ significantly from those of the electronic response. The tensor properties of the third-order material susceptibility associated with the fast electronic nonlinearity are well known.[1] For the radial acoustic mode approximation used in this study the electrostrictive material response is a scalar change in density, and we can express the electrostrictive contribution to one Cartesian component of the nonlinear polarization as

$$P_i^{(\mathrm{str})}(\omega_s) = 3\chi^{(\mathrm{str})} \sum_j |E_j(\omega_p)|^2 E_i(\omega_s)\,, \tag{5}$$

where $\chi^{(\mathrm{str})} \equiv \chi^{(\mathrm{str})}(\omega_s = \omega_s + \omega_p - \omega_p)$. This expression is valid for two interacting fields, where the pump at frequency ω_p is assumed to be much more intense than the signal at frequency ω_s and where the pump intensity does not vary significantly over the electrostrictive response time. Note that the degeneracy factor has the same value whether or not $\omega_s = \omega_p$; that is, the weak-wave retardation factor is equal to 1 for electrostriction.

We next introduce the fractional electrostrictive contribution $\eta \equiv n_2(\mathrm{str})/n_2(\mathrm{fast}) = \chi^{(\mathrm{str})}/\chi^{(3)}$. Here $\chi^{(3)} \equiv \chi_{iiii}^{(3)}$ includes both the electronic and the Raman contributions. When we retain only the automatically phase-matched terms, we arrive at an expression for the total nonlinear material polarization driving the nonlinear refractive index as the sum of electrostrictive and fast contributions. The tensor properties of the Raman susceptibility are taken here to be equivalent to the nonresonant electronic. For the SPM case $\omega_s = \omega_p$, we obtain

$$P_x^{\mathrm{NL}}(\omega_p) = 3\chi^{(3)}(1+\eta) \times \left[|E_x(\omega_p)|^2 + \frac{(2/3+\eta)}{(1+\eta)} |E_y(\omega_p)|^2 \right] E_x(\omega_p)\,, \tag{6}$$

whereas for the case of XPM, $\omega_s \neq \omega_p$, we obtain

$$P_x^{\mathrm{NL}}(\omega_s) = 6\chi^{(3)}(1+\eta/2) \times \left[|E_x(\omega_p)|^2 + \frac{(1/3+\eta/2)}{(1+\eta/2)} |E_y(\omega_p)|^2 \right] E_x(\omega_s)\,. \tag{7}$$

The contribution to the nonlinear refractive index from each polarization component of the pump can be expressed as $n_2(\mathrm{eff}) \equiv \kappa_p n_2(\mathrm{fast})$, where $n_2(\mathrm{fast}) = 12\pi^2\chi^{(3)}/n^2 c$ is the SPM index for linearly polarized light. The subscript p in the effective nonlinearity factor κ_p refers to the polarization state of the pump with respect to the probe: for the pump component parallel to the probe $\kappa_p = \kappa_{\|}$, and for orthogonal pump and probe $\kappa_p = \kappa_{\perp}$. If the polarization state is not maintained through propagation, the signal experiences an effective nonlinear phase shift proportional to the energy-weighted path average of the nonlinear response induced by the two orthogonal pump components.[12] At any position along the fiber the nonlinear index changes experienced by the x and y polarization components of the signal are

$$\Delta n_x = n_2(\mathrm{fast}) I_0[\kappa_{\|} f + \kappa_{\perp}(1-f)]\,, \tag{8a}$$

$$\Delta n_y = n_2(\mathrm{fast}) I_0[\kappa_{\|}(1-f) + \kappa_{\perp} f]\,, \tag{8b}$$

where a fraction $f = f(z)$ of the pump energy resides in polarization state x and a fraction $(1-f)$ in orthogonal state y. Because of the random nature of the polarization evolution we cannot determine explicitly the energy-distribution function $f(z)$. Instead, we introduce a probability distribution $p(f)$ describing the distribution of energy in polarization state x along

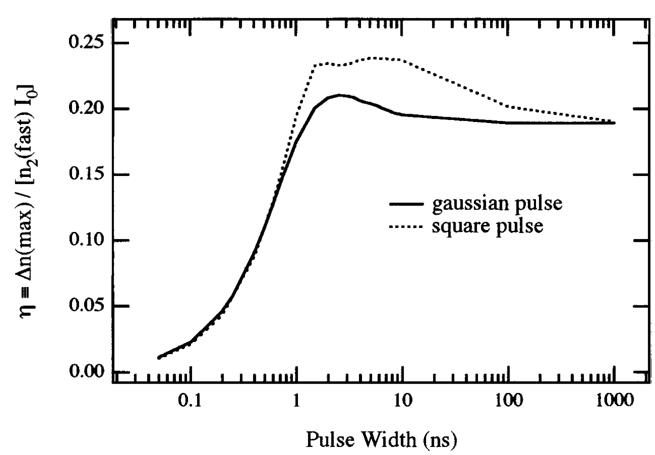

Fig. 1. Magnitude of the electrostrictive nonlinearity $\Delta n(\mathrm{max})$ relative to the fast nonresonant electronic contribution for linearly polarized light as a function of pulse width (FWHM) for a fiber with mode field radius $a = 4.5\ \mu\mathrm{m}$ and $n_2(\mathrm{fast}) = 2.96 \times 10^{-16}\ \mathrm{cm}^2/\mathrm{W}$.

Table 1. Effective Nonlinearity Factor

$\kappa_p = n_2(\text{eff})/n_2(\text{fast})$	SPM[a]	XPM
$\kappa_{\parallel}$	$1 + \eta$	$2\left(1 + \frac{\eta}{2}\right)$
$\kappa_{\perp}$	$\frac{2}{3} + \eta$	$2\left(\frac{1}{3} + \frac{\eta}{2}\right)$
κ_{eff} : random polarizations	$\frac{8}{9} + \eta$	$2\left(\frac{7}{9} + \frac{\eta}{2}\right)$
κ_{eff} : unpolarized	$\frac{5}{6} + \eta$	$2\left(\frac{2}{3} + \frac{\eta}{2}\right)$

[a]SPM and XPM refer, respectively, to the single- and dual-frequency situations, and $\eta = n_2(\text{str})/n_2(\text{fast})$.

the length of the fiber. An effective nonlinear change in the refractive index can then be expressed as the probability-weighted average of the local nonlinear response, that is, by

$$\begin{aligned}\langle \Delta n \rangle &= \int_0^1 [\Delta n_x f + \Delta n_y (1-f)] p(f) \mathrm{d}f \\ &\equiv \kappa_{\text{eff}} n_2(\text{fast}) I_0 .\end{aligned} \tag{9}$$

A study of the random walk associated with polarization evolution[12] suggests a uniform distribution for $f(z)$ such that $p(f) = 1$. Through simple integration of Eq. (9) that uses substitutions from Eqs. (8) and by use of $p(f) = 1$, the effective nonlinearity factor associated with propagation in a typical single-mode fiber is

$$\kappa_{\text{eff}}(\text{random pol.}) = (2\kappa_{\parallel} + \kappa_{\perp})/3 . \tag{10}$$

An exception arises when the pump or the probe is unpolarized, as in the XPM experiment discussed below. In this case there is no propagation dependence to the state of polarization; a local ensemble average always yields equal energies in any two orthogonal polarizations. This implies that $p(f) = \delta(f - 1/2)$ and

$$\kappa_{\text{eff}}(\text{unpol.}) = (\kappa_{\parallel} + \kappa_{\perp})/2 . \tag{11}$$

Through the use of Eqs. (6)–(11) we have calculated the effective nonlinearity factors κ_p for nonlinear interactions involving the fast nonresonant electronic and the steady-state electrostrictive responses. These results are summarized in Table 1. This procedure successfully reproduces the factor of $\kappa_{\text{eff}} = 8/9$ routinely cited for SPM in non-polarization-maintaining fiber[10,11] in the absence of electrostriction and the factor of $\kappa_{\text{eff}} = 2/3$ for XPM when unpolarized light is used.[6] A new result is the factor of $\kappa_{\text{eff}} = 7/9$ for the action of XPM in a system with randomly evolving polarization; this prediction remains to be confirmed by either experiment or numerical simulation.

We confirm the influence of the electrostrictive nonlinearity by examining the results of a recent XPM experiment that uses a harmonically modulated cw pump.[6] A pump with peak intensity I_0, modulation frequency Ω_0, and modulation depth m impresses a time-dependent nonlinear phase given by

$$\begin{aligned}\Delta\phi^{\text{NL}}(t) = \frac{2\pi}{\lambda} L \frac{m}{m+1} \{&2n_2(\text{fast}) \\ &+ n_2(\text{str}) \operatorname{Re}[H(\Omega_0)]\} I_0 \cos(\Omega_0 t + \varphi)\end{aligned} \tag{12}$$

onto a collinearly polarized probe at wavelength λ. The propagation distance is L, and φ is a small phase shift that arises from the imaginary part of the acoustic frequency response. In Ref. 6 the authors measured the nonlinear phase shift in dispersion-shifted fibers, using 7-MHz modulation of unpolarized pump light. They reported a value of the SPM index, adjusted to linearly polarized light for purposes of reference, of $n_2^{\parallel}(\text{reported}) = 3.35 \times 10^{-16}\ \text{cm}^2/\text{W}$. This value is to be compared with the value of $n_2^{\parallel}(\text{fast}) = 2.96 \times 10^{-16}\ \text{cm}^2/\text{W}$ obtained in mode-locked-pulse SPM experiments[3] for similar fibers, also adjusted for linearly polarized light. Using the factors of Table 1 relevant to unpolarized light, and recognizing the assumption of zero electrostrictive contribution implicit in the value reported in Ref. 6, we deduce that the relative electrostrictive contribution to the nonlinearity measured in this experiment is given by

$$\eta = 4\{[n_2^{\parallel}(\text{reported})/n_2^{\parallel}(\text{fast})] - 1\}/3 = 0.176 . \tag{13}$$

This 17.6% addition to the nonlinear refractive index is in excellent agreement with the expected electrostrictive response to a low-frequency modulation of the pump, as predicted by Eq. (3).

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