Measurement of the nonlinear optical response of optical fiber materials by use of spectrally resolved two-beam coupling

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We present measurements of the femtosecond nonlinear response of fiber waveguide materials, obtained with spectrally resolved two-beam coupling. This simple but sensitive technique provides all the parameters that characterize the third-order nonlinear optical response, including the dynamics. Measurements are made with nanojoule-energy pulses from a mode-locked Ti:sapphire laser and a few millimeters of waveguide preform, without the need for drawing fiber. © 1999 Optical Society of America

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Nonlinear optical phenomena in fused silica have received attention as a source of potential limitations to high-bit-rate transmission systems. Although most studies focus only on the magnitude of the nonlinear index of refraction n_2, the dynamics of the nonlinear response are equally important. Approximately 18% of the nonlinear index of refraction in fused silica is attributable to the Raman effect.[1] This nuclear contribution, with vibrational frequencies of up to $\sim 1200\,\mathrm{cm}^{-1}$, constitutes a noninstantaneous nonlinear response, which should be observable in the femtosecond regime. Stolen and Tomlinson investigated its effect on soliton formation and concluded that the effective n_2 will decrease with decreasing pulse duration.[2] This decrease will adversely affect transmission systems based on solitons. For all transmission systems the magnitude of n_2 is an important design parameter. With terabit-per-second rates already demonstrated, the dynamics of the nonlinear response will only assume greater importance in the future.

Waveguide manufacturers are currently working to develop fibers with nonlinearities that are smaller than those of the fibers in use at present, which are already among the least-nonlinear materials known. Development of such fibers poses the problem of reliably measuring such small nonlinearities in experimental, and therefore imperfect, materials. Although cw measurements of fiber nonlinearities offer the advantage of requiring only readily available diode pump sources with relatively low power,[3,4] these techniques cannot resolve the different contributions to the nonlinear index, such as electronic and nuclear nonlinearity, electrostriction, and thermal effects. Because of the low peak powers involved, hundreds of meters of fiber are typically required for generating a measurable nonlinear response. This requires high-quality fiber, which is often not available for experimental compositions. It is relatively easy to explore a wide range of glass compositions if only small samples are required, but considerable effort and expense are required for optimizing a composition for fiber-drawing properties. On the other hand, Ti:sapphire lasers that produce pulse durations of <25 fs are becoming widespread in research laboratories, and they recently became commercially available. We show here that, using a short-pulse laser and spectrally resolved two-beam coupling (SRTBC),[5] one can easily resolve the nonlinear optical response in a simple experiment by use of short pieces of waveguide preform, the precursor from which a fiber is drawn. The measurement provides the signs and the magnitudes of the real and the imaginary parts of the third-order susceptibility $\chi^{(3)}$, including their dynamics.

Present-day telecommunications fibers contain varying amounts of GeO_2 for the purpose of increasing the refractive index of the core above that of the cladding. Since the addition of GeO_2 to SiO_2 increases the nonlinear index of refraction in proportion to the germania content,[3] it is natural to question whether the Raman contribution also increases as well as whether its increase matches that of the total n_2. Here we investigate the dependence of the nonlinear response of SiO_2-GeO_2 glasses on germania content, using Raman spectroscopy and SRTBC. We find an increase in both the electronic and the nuclear parts of the response with GeO_2 content and a slight increase of the nuclear fraction.

The SRTBC experiment is a standard pump–probe experiment with the addition of a monochromator to select a narrow slice of the probe spectrum for observation. The change in refractive index that is induced by the strong pump pulse induces a nonlinear phase shift in the probe. For an instantaneous response this nonlinear phase shift follows the temporal envelope of the pump pulse, whereas for a noninstantaneous response time the phase shift will be the convolution of the response function and the temporal envelope of the pump pulse. Hence observation of the time-dependent nonlinear phase shift provides information about the material response function. Although it is possible to obtain the nonlinear response from two separate

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measurements of the absolute nonlinear phase shift and the absolute Raman cross section, these measurements are typically prone to large errors. With short pulses we obtain both contributions from a single measurement, thereby significantly reducing the error in the relative electronic and nuclear contributions.

It is possible to find closed-form analytical solutions for the SRTBC signal for pulses with a Gaussian temporal envelope and simple material response functions such as a weakly damped sinusoid[6] or an instantaneous response.[5] However, the envelope of an ultrashort laser pulse often does not have a simple analytic form, and neither does the Raman spectrum of waveguide material. In the most general case the SRTBC signal can be calculated numerically from the pulse spectrum and the Raman spectrum, which can both be easily measured.

The temporal envelope of the pulse is obtained from a zero-phase Fourier transform of the pulse spectrum, and the result is verified by comparison of the calculated and the measured autocorrelations. Assuming parallel polarizations for the pump and the probe beams, the nonlinear response function is obtained from the Raman cross section by use of[1]

$$\mathrm{Im}[\chi_{1111}^{\mathrm{vib}}(\Omega_{\mathrm{vib}})] \propto \frac{1}{\hbar\omega_0(\omega_0 - \Omega_{\mathrm{vib}})^3}\,\frac{\mathrm{d}^2\sigma_{\mathrm{hh}}}{\mathrm{d}\Omega\,\mathrm{d}\omega} \times [\exp(-\hbar\Omega_{\mathrm{vib}}/k_B T) - 1]. \quad (1)$$

Here $\mathrm{d}^2\sigma_{\mathrm{hh}}/\mathrm{d}\Omega\mathrm{d}\omega$ denotes the polarized ("horizontal–horizontal") differential scattering cross section, ω_0 is the laser frequency, and Ω_{vib} is the vibrational frequency. A Fourier transform of the imaginary part of the nonlinear susceptibility χ_{1111} with the additional condition that the response function must be real and causal yields the time-domain response function for the nuclear contribution.[7] The instantaneous electronic contribution can be added as a delta function, giving a total nonlinear response function

$$\Phi_{1111}(t - t_0) = n_2[(1 - f_n)\delta(t - t_0) + f_n R(t - t_0)], \quad (2)$$

where f_n is the fractional nuclear contribution. We do not attempt to measure the absolute value of the Raman cross section, since this calibration introduces a large uncertainty, but determine f_n from the time-domain signal. The time-dependent nonlinear phase shift $\Delta\varphi_{\mathrm{NL}}$ is obtained from a convolution of the response function and the envelope of the pump intensity I_{pu}:

$$\Delta\varphi_{\mathrm{NL}}(t) = A\int_{-\infty}^{t} \Phi_{1111}(t - t')I_{\mathrm{pu}}(t')\mathrm{d}t'. \quad (3)$$

The prefactor A depends on fundamental constants and the effective overlap between the pump and the probe pulses. Since the latter factor can be difficult to estimate accurately, it is most convenient to determine A from the signal produced by a reference material with known nonlinearity. For small nonlinear phase shifts, the probe field $E_{\mathrm{probe}}(t, \tau)$ after the nonlinear interaction depends on the input field $E_{\mathrm{probe},0}(t, \tau)$ and the nonlinear phase shift as

$$E_{\mathrm{probe}}(t, \tau) \approx E_{\mathrm{probe},0}(t, \tau)(1 + i\Delta\varphi_{\mathrm{NL}}), \quad (4)$$

where τ is the delay between the pump and the probe pulses. In our experiments $\Delta\varphi_{\mathrm{NL}} < 10^{-3}$ rad, so we disregard factors that are quadratic (or higher) in the nonlinear phase shift. After a Fourier transform, the signal S at detuning $\Delta\omega = \omega - \omega_0$ is

$$S(\Delta\omega, \tau) = \frac{|E_{\mathrm{probe}}(\Delta\omega, \tau)|^2 - |E_{\mathrm{probe},0}(\Delta\omega, \tau)|^2}{|E_{\mathrm{probe},0}(\Delta\omega, \tau)|^2}. \quad (5)$$

The result is linear in n_2 and f_n, which are the only free parameters. The shape of the SRTBC signal is highly sensitive to chirp,[8] so we take agreement between the calculated and the experimental SRTBC signals as confirmation that the pulses are transform limited.

Waveguide materials were prepared at Corning, Inc., by flame hydrolysis, by use of the outside vapor deposition technique. In this case a porous blank was initially formed, which was then consolidated at 1400–1500 °C in a helium atmosphere. Samples in the form of cubes of 5-mm length were cut from the consolidated preforms. Samples containing 0-, 8-, 19-, 36-, and 50-wt. % GeO_2 were measured by use of 18-fs pulses from a Ti:sapphire laser emitting at ~800 nm. The pump-pulse intensity was $\sim 20\,\mathrm{GW/cm^2}$ at the focus, and the pump and the probe had parallel polarizations. The most important telecommunication wavelength is 1.55 μm, but since the absorption edge of these materials lies in the ultraviolet, measurements made at 800 nm are already nonresonant and hence should closely approximate the nonlinear response at longer wavelengths. Preliminary measurements at 1.3 μm yield values for the n_2 of fused silica within the uncertainties of values obtained at 800 nm.

Figures 1(a) and 1(b) show SRTBC data obtained from samples with 0- and 50-wt. % GeO_2, respectively, along with the signals calculated as outlined above from the Raman spectra, shown in Fig. 2, and the laser spectrum. The bipolar part of the signal is due to the fast electronic response, and the damped oscillations seen for positive delay are due to the nuclear contribution. The calculated signals reproduce the measured data very well. We conclude that no significant dispersive broadening of the pulses occurred in the short (<1-mm) interaction region, as expected since the characteristic dispersion length is ~3 mm.

Results obtained from all the samples are summarized in Table 1. The total n_2 is given with respect to that of pure silica, which was determined to have a value of $(8.1 \pm 1.2) \times 10^{-14}$ esu by comparison with SF6 glass ($n_2 = 9.3 \times 10^{-13}$ esu), which agrees well with the value 7.4×10^{-14} esu determined by Boskovic *et al.*[3] Both the electronic and the nuclear contributions increase with the addition of GeO_2, but the nuclear contribution increases faster, leading to a slight increase in the nuclear fraction

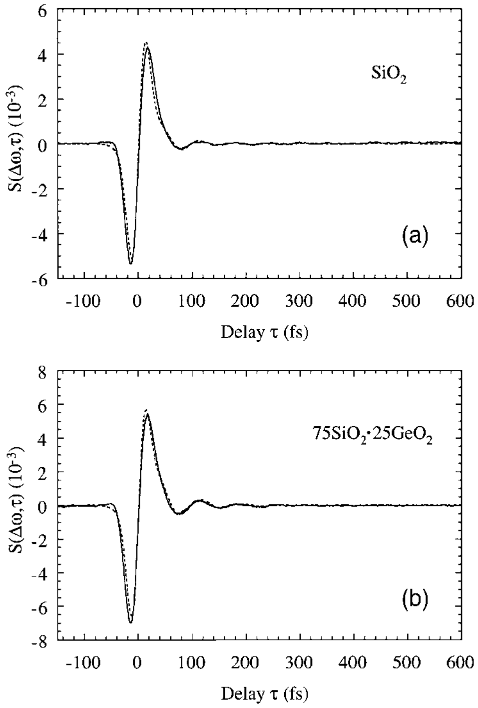

Fig. 1. SRTBC signal in (a) fused silica and (b) 75 SiO_2 · 25 GeO_2 (solid curves) and results calculated from spectra (dashed curves). The data were taken at a detuning $\Delta\omega = 0.7\sigma$, where σ is the FWHM of the spectrum.

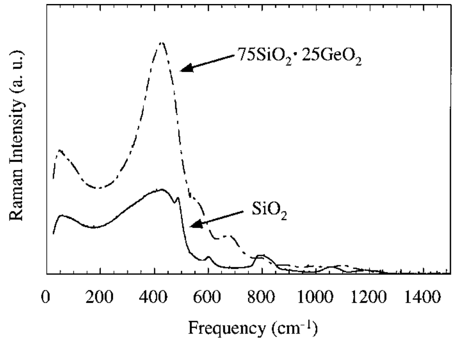

Fig. 2. Raman spectra for fused silica and 75 SiO_2 · 25 GeO_2.

f_n. For the sample with 50-wt. % GeO_2 we obtain $n_2 = (1.1 \pm 0.15) \times 10^{-13}$ esu, which agrees with the value $n_{2,\mathrm{calc}} = 1.1 \times 10^{-13}$esu, calculated by use of the empirical formula that Boskovic *et al.* obtained for 1.55 μm.[3] It is important to realize that the data of Boskovic *et al.* included an electrostrictive component of $\chi^{(3)}$, which Buckland and Boyd estimated contributes approximately 16% of the total nonlinearity at zero frequency.[9] This component does not contribute in our experiment, so the apparent exact agreement is coincidental. The good agreement validates our assumption that, for wavelengths beyond ~750 nm, n_2 is nearly independent of wavelength, and our results should be directly applicable to the telecommunications regime.

Table 1. Nonlinear Parameters of SiO_2–GeO_2 Glasses

GeO_2			
wt. %	mol. %	Relative n_2	Nuclear Fraction f_n
0	0	1.00	(13 ± 4)%
8	2.8	1.09 ± 0.11	(15 ± 4)%
19	7.3	1.13 ± 0.11	(16 ± 5)%
36	15.6	1.22 ± 0.12	(18 ± 5)%
50	25	1.39 ± 0.14	(17 ± 5)%

In conclusion, we have shown that spectrally resolved two-beam coupling by use of a short-pulse Ti:sapphire laser produces reliable measurements of the nonlinear response functions of waveguide materials. This simple but sensitive experiment, together with a straightforward analysis, can provide the nuclear and the electronic contributions to the optical nonlinearity of materials without the necessity for drawing them into fibers. In what we believe to be the first application of SRTBC to this problem, we found that the fractional nuclear contribution to the nonlinearity of SiO_2–GeO_2 glasses increases with germania content. We expect that this approach will be useful in the characterization of waveguide materials developed in the future.

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