Direct measurements of the nonlinear index of refraction of water at 815 and 407 nm using single-shot supercontinuum spectral interferometry

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Single-shot supercontinuum spectral interferometry was used to measure the nonlinear index of refraction due to the optical Kerr effect in water at both 815 and 407 nm, with pump pulse lengths of ~90 and ~250 fs, respectively. Knowledge of the nonlinear index at 407 nm allows pulse tailoring to achieve remote underwater pulse compression and self-focusing. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3142384]

The nonlinear index of refraction of water is a key physical parameter for any underwater use of high-powered lasers for which peak power is greater than the critical power, P_{crit}, for self-focusing. Such lasers include typical Q-switched lasers with pulse energies above a few millijoules, and ultrashort pulse Ti:sapphire systems with pulse energies above 1 nJ. Applications of water propagation of intense laser pulses include femtosecond laser surgery,[1] underwater femtosecond laser ablation,[2] and underwater femtosecond laser shock peening.[3] One potential application of particular interest is remote underwater pulse compression. Underwater transmission is optimal for blue-green wavelengths, where low linear absorption, e.g., $\alpha \sim 0.005\ \mathrm{m}^{-1}$ at 400 nm,[4] makes these shorter wavelengths much more suitable for long range nonlinear self-focusing. By combining nonlinear self-focusing with temporal compression due to group velocity dispersion of a prechirped broadband pulse, it is possible to attain intensities high enough to cause breakdown in water. Precise and accurate knowledge of the nonlinear index of refraction of water, n_2, which is in general a function of the optical frequency, as well as the optical pulse duration,[5] is necessary to model underwater pulse compression and to experimentally produce optical breakdown at predetermined remote locations.

In this letter, we report the demonstration of a time-resolved, profile-independent, and precise measurement of n_2 of water at 407 nm for a subpicosecond pulse duration. We first perform the experiment at 815 nm [21 nm full width at half maximum (FWHM) bandwidth] as a benchmark and proof of technique. Then a measurement at 407 nm (produced by frequency doubling with a 240 μm beta-barium borate (BBO) crystal, yielding 4 nm FWHM bandwidth) is made by modification of the setup. Our technique, single-shot supercontinuum spectral interferometry (SSSI),[6,7] provides high temporal resolution [~10 fs (Ref. 7)] and a wide observation window (~2 ps) for measurement of the nonlinear response of an optical medium. Such measurements can show whether or not the nonlinear response is nearly instantaneous, that is primarily due to nearly instantaneous and isotropic electronic response, or has delayed and possibly nonisotropic contributions from molecular vibrations and rotations.[8,9] Plasma contributions to the response can also be distinguished in SSSI measurements,[7] an especially important feature at the high intensities typical of ultrashort pulse lasers, and for nonlinear optical interactions in general. Many previously reported n_2 values were inferred from measurement techniques dependent on self-focusing dynamics.[10–13] Such techniques are highly dependent on the spatial profile of the laser pulse, as well as its reproducibility, and typically yield large errors, conflicting values for n_2,[5,11] or both. Additional published techniques for measuring n_2 utilized elliptical polarization rotation,[14] spatial profile analysis,[15] and spectral analysis of self-phase modulation.[16,17] Some reported values of n_2 for water differed from the majority by an order of magnitude,[12] leaving uncertainty about its true value. We believe these differences have been resolved by the present measurements. Finally, the excellent shot-to-shot stability of the kilohertz regenerative amplifier system used here, in combination with the averaging of hundreds of interferograms, allowed extraction of small phase shifts (~0.01 rad), resulting in a small n_2 measurement error.[8]

The pump-probe SSSI setup used was similar to the setup in Ref. 7, with a flowing water interaction cell replacing the gas cell. A 1 mJ, 1 kilohertz, Ti:sapphire laser with FWHM pulse length of 90 fs and central wavelength of 815 nm was used to generate all pulses required for the SSSI technique. The pulse width was reduced from the system's original 130 fs using acousto-optical pulse shaping in the spectral domain.[18] SSSI allows extraction of a space and time-domain phase shift, $\Delta\phi(x,t)$ from interferograms produced by the setup, where x and t correspond to the transverse spatial location (across the beam) and time, respectively. From $\Delta\phi(x,t)$, the change in refractive index $\Delta n(x,t)$, and thus n_2, caused by cross-phase modulation (XPM) from a pump beam can be obtained from,[7]

$$\Delta\phi(x,t) = \frac{\omega}{c}\int_0^z \Delta n(x,z',t)dz' = \frac{\omega}{c}\Delta n(x,t)L, \qquad (1)$$

where L is the effective interaction length as discussed below.

In the experiment, the laser output was split into two beams. One beam was used to generate twin supercontinuum (SC) pulses, reference followed by probe, each 2 ps long with ~100 nm bandwidth centered around 690 nm.[7] The other beam acted as the pump, either 815 or 407 nm, and was

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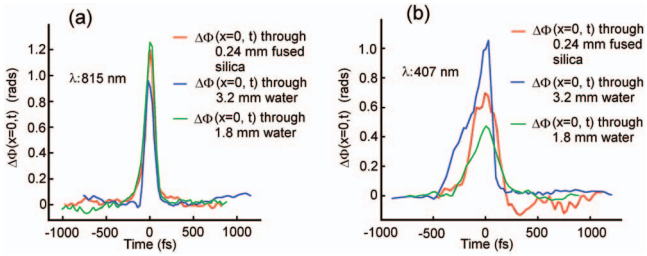

FIG. 1. (Color) (a) $\Delta\phi(t)$ for a 815 nm pump pulse through 0.24 mm fused silica, 3.2 mm water, and 1.8 mm water. The fused silica trace indicates the shape of the original pump pulse, and these plots allow comparison of pump profiles with those of the resulting $\Delta\phi(t)$. Pulse energies were varied to obtain similar final phase shifts. Pulse temporal profiles are the same, indicating no broadening or distortion. (b) $\Delta\phi(t)$ for a 407 nm pump pulse through 0.24 mm fused silica, 3.2 mm of water, and 1.8 mm of water. For the fused silica and 1.8 mm water propagation, pulse temporal profiles are very similar. For 3.2 mm of water propagation, the pulse temporal profile is slightly distorted, possibly by high order dispersion.

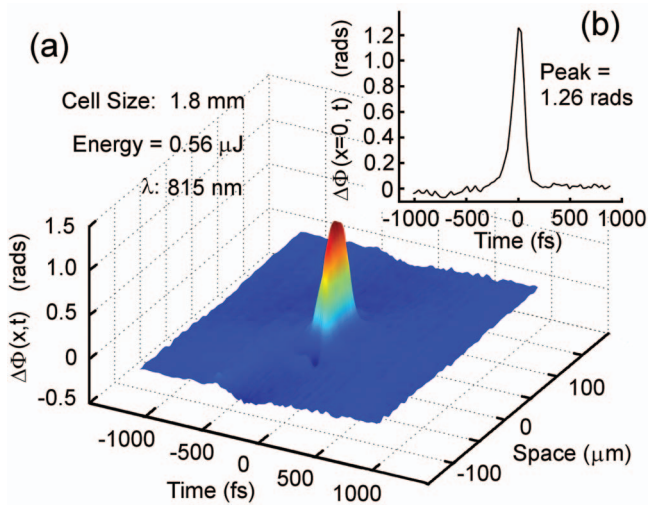

FIG. 2. (Color) (a) Space and time-resolved phase shift, $\Delta\phi(x,t)$ extracted from a 300 shot averaged interferogram. Pump pulse: 815 nm, 90 fs, $I_o = 1.9\times10^{11}$ W/cm^2; water cell thickness=1.8 mm. (b) Phase shift lineout $\Delta\phi(x=0,t)$ along the pump axis, $\Delta\phi(0,0)=1.26$ radians, yielding $n_2^{\mathrm{SPM}}(815\ \mathrm{nm}) = 1.9\times10^{-16}\pm10\%$ cm^2/W.

propagated collinearly and cotemporally with the probe SC pulse through the water cell interaction region. The water (doubly distilled fresh water at 71 °F) flow was sandwiched between two thin (120 μm) fused silica windows, chosen to minimize self-phase modulation of the pump and cross phase modulation of the probe, which could deleteriously contribute to the SSSI spectrum and phase. Propagating the pump pulses through the drained water cell verified no observable spectrum or phase shift contribution from the fused silica windows. Since nonlinear response in fused silica is very nearly instantaneous,[17] the temporal profile of the pump laser pulse could be obtained by raising its energy and observing its nonlinear phase shift during propagation solely through the fused silica windows. Figure 1(a) shows typical profiles of the 815 nm pump pulse after propagation through 0.24 mm of fused silica, 3.2 mm of water, and 1.8 mm of water. Figure 1(b) shows typical 407 nm phase shift profiles during propagation through 0.24 mm of fused silica (which shows the original pump profile), as well as for 3.2 and 1.8 mm of water. For 407 nm, the temporal profiles though the 1.8 mm water cell and fused silica were very similar. In the 3.2 mm water cell, we observed a slight distortion of $\Delta\phi(t)$. We suspect this distortion was caused purely by higher order dispersion of the pump pulse, as the pulse duration was not significantly increased, and a similar distortion was not observed for the 1.8 mm water cell. Since we did not observe any significant pulse broadening or shifting between $\Delta\phi(x,t)$ for water and fused silica for any case, we conclude that the nonlinear response of water is a near-instantaneous electronic response. Evidently, nonresonant Raman excitation of water molecules, which could give rise to a fast yet delayed response, play little role under the conditions of this experiment. We note that the previous techniques for measurement of nonlinearities[11–13] would not be able to make this assessment.

Based on an instantaneous response, the observed phase shift can be expressed as $\Delta\phi(x,t)=(\omega/c)\Delta n(x,t)L=(\omega/c)n_2I(x,t)L$, where n_2 is the nonlinear index of refraction, defined by $n=n_0+n_2I$. n_2 is a function of the third-order nonlinear susceptibility tensor, $\chi^{(3)}(-\omega_2;\omega_2,-\omega_1,\omega_1)$, where ω_1 and ω_2 are the pump and probe frequencies, respectively. It is important to note that when measuring n_2 with a pump-probe setup, the effect on the weak probe is twice as large as that for self-phase modulation (SPM), for which $\omega_1=\omega_2$.[5] In esu, $n_2^{\mathrm{SPM}}=3\pi\chi^{(3)}(-\omega_1;\omega_1,-\omega_1,\omega_1)/n_0$, and $n_2^{\mathrm{XPM}}=6\pi\chi^{(3)}\times(-\omega_2;\omega_2,-\omega_1,\omega_1)/n_0$, where n_0 is the linear index of refraction.[5] n_2 is then extracted from the measured phase shift $\Delta\phi$ and the interaction length, $L<2z_0$, where z_0 is the pump beam Rayleigh length. The interaction lengths L associated with the two water cells used in our experiment were 1.8 and 3.2 mm, each well below $2z_0$ for both the 815 nm ($2z_0=9.0$ mm) and 407 nm ($2z_0=6.4$ mm) pulses.

After the beams exited the water cell, the pump beam was removed with a normal incidence high reflectivity dielectric mirror, while the reference and probe beams continued through a lens which imaged the water cell exit plane onto the entrance slit of the spectrometer. A charge-coupled device (CCD) camera located at the spectrometer image plane recorded the interference of the pump and probe in the spectral domain, with spatial resolution along the entrance slit direction. For each parameter set (pump laser energy, pump wavelength, beam waist location, cell thickness), 300 interferogram images were taken and then averaged. Owing to the high shot-to-shot reproducibility of the kilohertz pump laser pulses, interferogram averaging resulted in a measurement noise level (from CCD pixel noise) much lower than that from a single shot.[8]

From Eq. (1), n_2^{SPM} was extracted from $\Delta\phi(x=0,t=0)=(\omega/c)n_2^{\mathrm{XPM}}I(x=0,t=0)L=(\omega/c)2n_2^{\mathrm{SPM}}I(x=0,t=0)$, requiring determination of $I(x=0,t=0)=I_o$. Using the measured pulse energy and temporal pulse profile [from Figs. 1(a) and 1(b)], a peak pulse power was calculated. Magnified CCD images of the pump spot at the interaction location were used to determine the transverse beam profile. Calibration of the CCD with respect to laser pulse energy provides the peak intensity for the given pulse length. We found $I_0\sim1\times10^{11}$ W/cm^2, well below the water ionization threshold of $\sim10^{13}$ W/cm^2 for 100 fs pulses.[19] Calculated peak powers were 3.3 MW for the 815 nm pump pulse and 1.8 MW for the 407 nm pump pulse, below the reported critical self focusing power of 4.4 MW for 800 nm.[11,13] This ensured that propagation was dominated by the external lens. Figure 2(a)

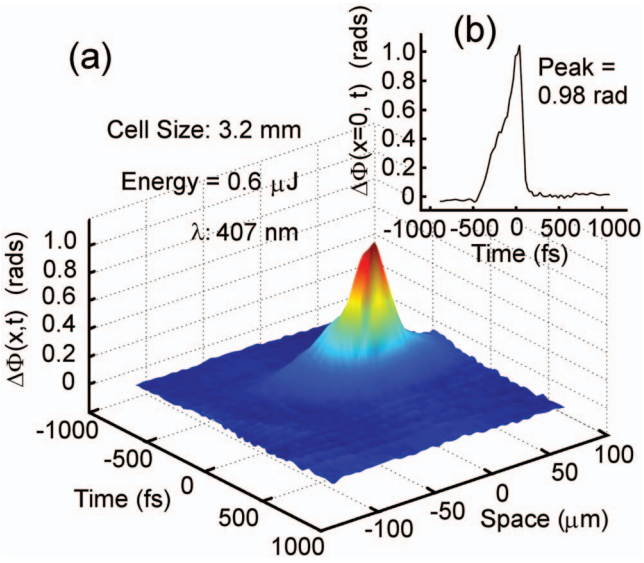

FIG. 3. (Color) (a) Space and time-resolved phase shift, $\Delta\phi(x,t)$ extracted from a 300 shot averaged interferogram. Pump pulse: 407 nm, 250 fs, $I_o = 9.4\times10^{10}$ W/cm^2; water cell thickness=3.2 mm. (b) Lineout along the pump beam axis, $\Delta\phi(x=0,t)$. The peak phase shift is $\Delta\phi(x=0,t=0) = 0.98$ rad, yielding $n_2^{\mathrm{SPM}}(407\ \mathrm{nm}) = 1.7\times10^{-16}\pm12\%$ cm^2/W.

shows $\Delta\phi(x,t)$ for the 90 fs FWHM, 815 nm pump pulse, with polarization parallel to the SC probe. Figure 2(b) shows an on-axis lineout with a peak phase shift $\Delta\phi(0,0)\sim1.26$ radians. In Fig. 2(a), no features or resurgent peaks appear in time after the pulse, indicating no effects from molecular rotation.[8]

Using the 815 nm peak intensity, measured as described above, an effective n_2 for SPM was determined to be $1.9\times10^{-16}\pm10\%$ cm^2/W, where the error is due to uncertainty in calibration of the photodiode used to measure pulse energies. This value represents an effective n_2 which an 815 nm pulse would experience as SPM. The linear dependence of $\Delta\phi(0,0)$ on peak intensity, I_o, was checked by comparing shots of different energy. Our measured n_2 value is significantly lower than the 2.7×10^{-16} cm^2/W reported in Ref. 20. Using our value to calculate the critical nonlinear self-focusing power via Eq. (2) below, yields $P_{\mathrm{crit}}=3.9$ *MW*. This corresponds well to the critical self-focusing power reported in Refs. 11 and 13 of 4.4 MW,

$$P_{\mathrm{crit}} = \frac{3.77\lambda^2}{8\pi n_o n_2}. \tag{2}$$

Additionally, perpendicular polarization between pump and probe was studied. For the SC probe polarized perpendicularly to the pump, the measured phase shift was reduced by a factor of 3, owing to the symmetry of $\chi^{(3)}$ in an isotropic medium,[5] and leaving the extracted n_2 unchanged.

Figure 3(a) shows $\Delta\phi(x,t)$ from the 407 nm pump pulse, peak intensity $I_o=9.4\times10^{10}$ W/cm^2 and pulse length ~250 fs FWHM (determined using the high energy nonlinear phase shift in fused silica windows, as previously described) through a 3.2 mm water cell. An on-axis lineout of the phase shift, $\Delta\phi(x=0,t)$, Fig. 3(b), gives $\Delta\phi(0,0)=0.98$ radians. This gives $n_2^{\mathrm{SPM}}(407\ \mathrm{nm}) = 1.7\times10^{-16}\pm12\%$ cm^2/W, where again the error is due to uncertainty in calibration of the photodiode used to measure pulse energies. As for 815 nm, the linearity of $\Delta\phi(0,0)$ with I_{peak} was checked by comparing 407 nm shots of different energies. The calculated n_2 corresponds to a critical self-focusing power, $P_{\mathrm{crit}}=1.1$ MW. As in the 815 nm case, a perpendicularly polarized probe experienced a factor of 3 reduction in phase shift, in agreement with theory,[5] and yielding the same value for n_2^{SPM}. The same value for n_2^{SPM} was also measured for the $L=1.8$ mm cell.

In conclusion, we have directly measured, using SSSI, the nonlinear index of refraction for distilled water at both 815 and 407 nm for subpicosecond pulses. Our measured values of n_2 agree with some previous indirect measurements at 815 nm. The present measurement technique allowed for observation of possible pump-induced after effects in the refractive index during a 2 ps time window, of which none were observed. Our directly measured value for n_2 of water at 407 nm has much less uncertainty than values extrapolated from measurements at other wavelengths and pulse durations, using less precise techniques. Values of n_2 reported here are expected to also pertain to sea water.

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