Anomalous laser-induced group velocity dispersion in fused silica

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Abstract: We present 20fs^2 accuracy laser-induced group velocity dispersion (LI-GVD) measurements, resulting from propagation of a femtosecond laser pulse in 1mm of fused silica, as a function of peak intensity. For a 5.5×10^{11} W/cm^2 peak intensity, LI-GVD values are found to vary from −3 to + 15 times the material GVD. Normal induced dispersion can be explained by the Kerr effect, but anomalous LI-GVD, found when the input pulses have negative pre-chirp, cannot. These findings have significant implications regarding self-compression and the design of femtosecond lasers.

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1. Introduction

To first approximation, propagation of femtosecond lasers in transparent media leads to pulse broadening due to dispersion. For visible and near-IR wavelengths pulses experience positive group delay dispersion (GDD) or normal dispersion, which is proportional to the length of the medium. In general, group velocity dispersion (GVD) corresponds to GDD per unit length, often quoted for ultrafast optics in fs^2/mm. Since the early days of ultrafast lasers the static or intensity independent GVD values for common optical materials were tabulated and methods were introduced to compensate the dispersion at least to second and third order. Laser induced changes to the refractive properties of the medium resulting in frequency broadening were first observed 45 years ago [1]. However, despite all the progress in nonlinear optics during the past decades, one can still expect to find surprising phenomena, especially as ultrashort femtosecond pulses with dispersion control to third-, fourth-, and even higher-orders become widely available.

When a high intensity laser pulse enters a dielectric medium, it perturbs the medium, causing nonlinear changes of the index of refraction leading to free carrier excitation [2–5], and defect generation [6,7]. These changes in turn affect the pulse characteristics as discussed in self-steepening [2]. Peak intensities of the order of 10^{11} W/cm^2 are below the threshold for supercontinuum generation, but are high enough to induce optical Kerr effects in fused silica [8]. Kerr effects produce an intensity-dependent refractive index change, which has been measured with high sensitivity and time resolution in a number of media [9–11]. Changes in the refractive index give rise to self-phase modulation (SPM). SPM causes frequency chirp and the generation of additional frequency components. It has been suggested that gas-phase molecular dynamics can lead to deterministic changes in the refractive index of the medium, as demonstrated experimentally by taking advantage of anomalous GDD associated with a rotational wave packet to compress laser pulses [12]. Soliton pulse compression, associated with self-compressed optical filaments caused by SPM have been observed and associated with the gas-glass-gas interfaces [13].

In general SPM affects femtosecond laser performance. In femtosecond lasers, nonlinear propagation effects such as SPM are therefore mitigated by temporal stretching of the pulses, as in chirped pulse amplification, or spatially expanding the laser beam, as in large mode area fibers. In ultrafast fiber laser design, high-energy pulses can accumulate a non-linear phase, causing wave-breaking and temporal profile changes of the output pulses [14]. Despite nonlinearity mitigating approaches, day-to-day variations in the performance of femtosecond lasers are quite common. It is therefore of fundamental importance to search for and quantify directly laser induced GVD and to provide a physical understanding that can be used for designing and modeling ultrafast lasers and devices. Our ability to measure and control spectral phase changes caused by nonlinear propagation in the spectral domain with high sensitivity [15], provides a new window into nonlinear optical processes. Here, we take a closer look at the second derivative of the spectral phase or GDD acquired by high peak-intensity laser pulses during propagation in fused silica. We explore transform limited pulses and pulses for which a positive or negative pre-chirp is impressed before propagation in fused silica. We associate the observed GDD changes with laser-induced GVD of the medium.

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2. Experimental setup

A schematic of the experimental setup is presented in Fig. 1. The output from a Ti:Sapphire oscillator (Coherent Inc.) passes through a pulse shaper (femtoFit, BioPhotonic Solutions, Inc.) and a regenerative amplifier (800 nm central wavelength, 1 kHz rep. rate, Legend, Coherent Inc.) and, after attenuation, is focused with a long focal length lens (f = 300 mm). The laser pulses are first characterized and high-order dispersion is eliminated to better than fifth order at the sample by using multiphoton intrapulse interference phase scan (MIIPS) [15]. The initial chirp of the pulse is controlled by the same pulse shaper. The transform limited pulse duration at full-width half-maximum (FWHM) is 40 fs. For all the measurements presented here the average power is set to 10 mW; low enough to prevent unwanted nonlinear processes away from the sample. The sample, a 1 mm plate of fused silica, is placed on a translation stage. The peak intensity was varied by scanning the sample position along the converging laser beam (from 56 to 8mm from the focal plane), in a manner similar to a Z scan [11], however the sample is not scanned through the focus. The focusing beam characteristics were measured using a beam profiler (LaserCam-HR, Coherent Inc.).

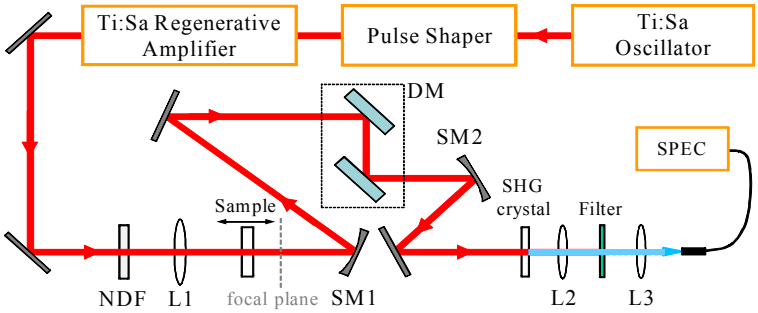

Fig. 1. Experimental setup for direct GDD measurements. NDF, neutral density filter; L1,2,3, lenses; SM1, 2, spherical mirrors; DM, chirped dielectric mirrors; SPEC, spectrometer.

The laser beam, collimated after the sample, is guided through a pair of chirped dielectric mirrors which introduce significant -1×10^5 fs^3 third-order spectral phase. After focusing on a 0.1-mm-thick BBO crystal the second harmonic generation (SHG) signal is recorded with a fiber-coupled compact spectrometer (Ocean Optics, HR4000). The SHG crystal is set far from the focal plane to avoid unwanted nonlinear processes and minimize beam profile effects. Reflective optics between the sample and the SHG crystal minimize spectral phase distortions.

For fused silica at 800nm, the GVD is equal to 36.2 fs^2/mm. Third order dispersion, from the dielectric mirrors, causes a narrowing of the SHG spectrum due to nonlinear optical interference, as discussed in terms of multiphoton intrapulse interference [16]. When GDD is added to a pulse with third-order spectral phase, the peak SHG wavelength shifts [16]. Here, GDD was measured by tracking the wavelength shift as shown in Fig. 2. The spectrometer resolution 0.12 nm was improved by using a Gaussian fit of the experimental spectra to determine the peak position, thus achieving ± 5 fs^2 precision. A calibration curve was made by scanning a known value of GDD using the interferometrically calibrated pulse shaper. Measuring GDD by tracking the spectral shift provides single-shot high-accuracy values (± 20 fs^2), without the need for phase retrieval algorithms. We further tested the absolute accuracy by measuring the dispersion from different thicknesses of fused silica; fitting the obtained values resulted in a slope of 37.6fs^2/mm, which is off by 1.4 fs^2/mm from the literature value. All measurements presented in this paper were obtained using this method, which we call real-time MIIPS (RT-MIIPS) [17]. A typical GDD measurement as a function of peak intensity involved 2500 measurements which were completed in less than 5 minutes.

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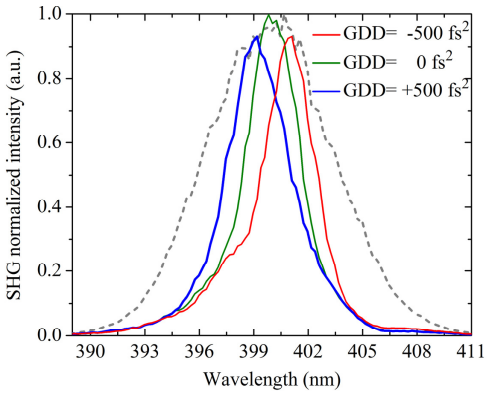

Fig. 2. RT-MIIPS principle. Experimental SHG spectra for different GDD values. The SHG spectrum for transform limited pulse is shown as a dotted line for reference. Cubic spectral phase causes SHG spectral narrowing (green). The SHG peak value shifts towards longer (shorter) wavelengths in the presence of negative (positive) GDD, respectively.

3. Results and discussion

The peak intensities, measured before the sample, ranging from 0.5×10^{11} W/cm^2 to 5.8×10^{11} W/cm^2, cover an important region where spectral changes occurring through SPM are small. Measured GDD values for a 1 mm fused silica plate as a function of peak intensity for five different pre-chirp conditions are presented in Fig. 3(a). The dispersion length L_D of fused silica is ~44 mm, and the nonlinear length L_{NL} at 5×10^{11} W/cm^2 is ~1mm. Therefore no significant dispersion or nonlinear phase shift from propagation through the sample are expected. Our measurements, however, indicate GDD changes are very significant despite the fact that the spectrum of the laser before and after the sample for intensities below 4×10^{11} W/cm^2 shows no difference or shift, for higher intensities one can notice ~5% wings resulting from SPM.

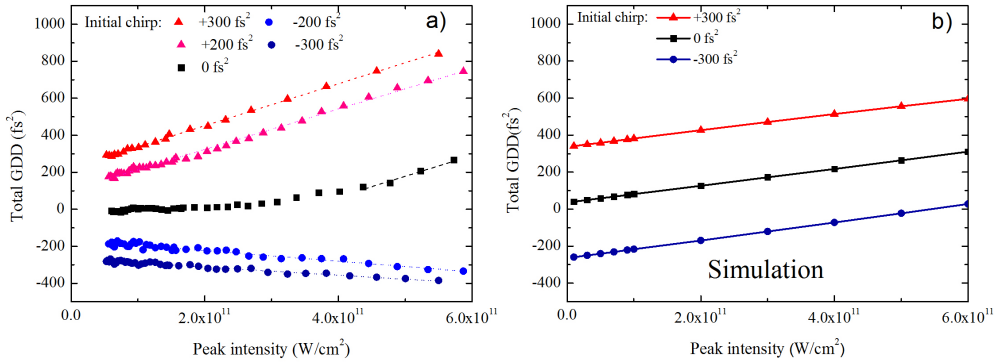

Fig. 3. Group delay dispersion as a function of peak intensity after propagation through 1 mm of fused silica. (a) Experimental measurements. The applied initial pre-chirps are + 300 fs^2 (red triangles), + 200 fs^2 (pink triangles), 0 fs^2 (black squares), –200 fs^2 (blue circles), and –300 fs^2 (dark blue circles). Dotted lines are corresponding linear fits. (b) Numerical, split-step Fourier transform, simulation without adjusting parameters. Initial pre-chirps are + 300 fs^2 (red triangles), 0 fs^2 (black squares), –300 fs^2 (dark blue circles). Note that the simulation fails to predict anomalous laser induced GVD for negatively pre-chirped pulses.

The most interesting observation in Fig. 3(a) is that the induced dispersion has a different sign depending on the initial chirp impressed on the input pulses. We define induced GDD as the measured GDD minus the initial chirp and static dispersion from the material. Positive pre-chirped pulses acquire normal induced GDD that is linear with peak intensity. Induced

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GDD measured at 5.5×10^{11} W/cm^2 is about 550 fs^2 and can be understood in terms of SPM. Induced GDD for the TL pulses increases slowly up to 4.5×10^{11} W/cm^2 followed later by a linear dependence. At an intensity of 5.8×10^{11} W/cm^2 the induced GDD for transform limited pulses reaches 270 fs^2. The dependence at lower intensities does not show a linear behavior, but the region from 4.5×10^{11} W/cm^2 to 5.8×10^{11} W/cm^2 can be fitted by a line (black dashed line in Fig. 3(a)) with a slope close to the slope that is observed for the positively pre-chirped pulses. The measurements with transform limited pulses appear to involve an intensity threshold before LI-GVD takes place. For negatively pre-chirped pulses an unexpected behavior is observed. Induced GDD is found to have a negative slope (blue dashed line in Fig. 3(a)) and continues to accumulate anomalous induced GVD, reaching a value of −120 fs^2 for 5.5×10^{11} W/cm^2. Effective nonlinear n_2 coefficients may be estimated based on fitting the slope of the experimental data, and using the approximate relation valid for low intensities:

$$n_2^{eff} \approx \frac{\varphi''(\omega-\omega_0)\cdot c}{2\tau_0^2\omega_0 L\cdot I_{PEAK}},$$

where the induced GDD, or the value of second derivative of the phase $\varphi''(\omega-\omega_0)$, is multiplied by the speed of light c, divided by the pulse duration τ_0, the angular frequency ω_0, the propagation length L and the peak intensity I_{PEAK}. The effective n_2 for positive and zero chirp is $n_2^{\mathrm{eff}} \approx (1.2 \pm 0.1) \times 10^{-16}$ cm^2/W, which is close to the published n_2 for fused silica $(2.48 \pm 0.23) \times 10^{-16}$ cm^2/W, measured with 100 fs pulses at 804 nm [18]. For negatively pre-chirped pulses the effective nonlinear refractive indices extracted are negative and about one quarter of the effective nonlinear refractive index obtained for positively pre-chirped pulses. To our knowledge, anomalous laser-induced GVD has not been observed before. Our measurements starting below 10^{11} W/cm^2 reveal a linear behavior observed up to 6×10^{11} W/cm^2. The linear trend for normal and anomalous LI-GVD, which starts at the lowest peak intensities, implies that free-carriers are not likely involved at these intensities.

We have carried out split-step Fourier transform numerical calculations to simulate our data [19]. Results for three initial chirp conditions are shown in Fig. 3(b). Our simulations, without adjustable parameters, agree with the findings for positive pre-chirp and those for TL pulses. The difference in the value of the slope is likely due to the intensity distribution of the laser in the experiment and in the calculations. However, the calculations do not agree with the experimental findings for negatively pre-chirped pulses. The observation of anomalous dispersion associated with negative pre-chirp requires us to consider a different nonlinear process. Smolorz et al. considered two-beam coupling for chirped pulses [20], and indicate energy is transferred from high-frequency to low frequency components during the interaction. Energy transfer depends on phase matching and depends on the relative time delay between the chirped pulses. The relevance of beam coupling to our single-beam experiment is that only a negatively chirped pulse can experience energy transfer caused by the non-instantaneous response of the medium, from high-frequencies to low frequencies. This energy transfer may be responsible for the observed anomalous LI-GVD. At higher intensities, once significant SPM related spectral broadening is observed (data not shown), the anomalous LI-GVD trend is reversed.

4. Conclusion

We have measured laser-induced group velocity dispersion in fused silica with high accuracy in a regime that is near the transform limit. The experiments were conducted using a direct single-shot measurement of dispersion. We have shown that LI-GVD depends linearly on the peak power of the laser and it can be in excess of one order of magnitude the static GVD of the material. Most significantly, anomalous LI-GVD is observed when the input pulses are

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negatively pre-chirped. Normal laser induced dispersion can be understood in terms of self-phase modulation, however, anomalous LI-GVD cannot. We speculate anomalous LI-GVD is caused by energy transfer from higher to lower frequencies. Our findings could have important implications for femtosecond laser design and compression, as well as for ultrafast laser science where knowledge of the exact value of frequency chirp is required.

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