Simultaneous determination of the materials' nonlinear refraction and ultrashort pulse properties by single-beam nonlinear ellipse signal

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Abstract

We proposed a single-beam method based on the nonlinear-ellipse-rotation measurement in thick samples which is able to determine simultaneously the samples' third-order nonlinearities and the pulses parameters. In this case, the nonlinear materials' properties can be determined more precisely since all measurements are carry out at the same laser conditions.

Nowadays, ultrashort laser pulses are important for nonlinear materials' characterization due to their high intensities. However, in comparison to long pulses, they are more complex in terms of the electromagnetic phase and amplitude, and to be able to measure the correct nonlinear optical properties, we must be more careful with the pulse propagation, dispersion of the materials employed in the optical components, etc. As it is known, the large bandwidth of ultrashort pulses affects the pulse width as the pulse propagates along a sample due to the materials' chromatic dispersion. This dispersion, which affects the pulse intensity, can be described by a frequency-dependence change φ in the spectral phase. In general, the most important term, which affects the pulse width and, consequently, the pulse intensity, is the second derivative ($\partial^2\varphi/\partial\omega^2=\varphi''$), or the group delay dispersion (GDD), given in fs^2, for instance. The maximum laser intensity occurs when the pulse is the shortest or transform-limited (TL), and the GDD is near zero. For well-behaved pulses where GDD is the most relevant term, it is possible to achieve near TL pulses by using appropriate pulse compressors, but this is not usual when a tunable frequency source like an optical parametric amplifier (OPA) is employed. In this way, it is interesting to measure the laser pulse parameters using suitable pulse characterization techniques such as FROG, SPIDER, and autocorrelation (AC) [1], for instance. Indeed, it is important to know the pulse parameters, particularly at the sample position where the nonlinear optical properties of the materials are measured. However, these pulse characterization devices are usually assembled separately in other laser beam lines. Here, we propose a simple single-beam method for measuring both the materials' third-order nonlinear optical refraction, n_2, and the pulse parameters (chirp and pulse width) by the nonlinear ellipse rotation (NER) measurements in thick samples [2] where the local nonlinearities are measured. This method uses the positive GDD of the optical materials to expose the pulse chirp and width. Basically, the nonlinear sample's GDD itself changes the pulse width, according to the input pulse chirp, as it propagates along the material. For example, if the input pulse chirp is negative (positive), the NER signal increases (decreases) as the focal point moves inside the nonlinear sample.

For sake of simplicity, we considered only the second-order and ignored other high-order dispersions, and, for a TL Gaussian pulse with duration τ_0 (FWHM) experiencing GDD of magnitude D_2, the pulse duration always increases according to [3]:

$$\tau = \tau_0 \sqrt{1+\left(4ln2\frac{D_2}{\tau_0^2}\right)^2}\ . \qquad (1)$$

The magnitude of D_2 is given by fs^2 and τ_0 in fs, for instance. In this case, the dispersion of the nonlinear sample needs to be known, and the dispersion as a function of propagation is:

$$D_2 = D_{2,pulse} + V_{2,sample} * L\ , \qquad (2)$$

where L is the focal point distance to the input interface, $D_{2,\mathrm{pulse}}$ is the chirp of the incident pulse at the sample entrance, and $V_{2,\mathrm{sample}}$ is the material's group velocity dispersion (GVD). Our method using thick samples works since the NER signal, which is proportional to the laser irradiance and the nonlinearity, changes along the sample's position.

In order to test our method, we have measured SF6 and LaSF-N30 optical glasses (6 mm) using a *Z*-scan setup with a lens with a 3 cm focal length, ultrafast laser pulses from a Ti:sapphire amplified laser system (Dragon, λ=780 nm, 1 kHz repetition rate, τ_0~40 fs, from K&M Labs), and a tunable source: OPA (Topas Prime, Light Conversion). Both SF6 and LaSF-N30 are adequate materials for testing due to their high nonlinear refraction and high dispersion, which are determined easily by their Sellmeier linear index of refraction curves. As can be seen in Fig. 1 in SF6, measuring the NER signals for different input pulses with different GDD, the signal changes according to Eq. 1.

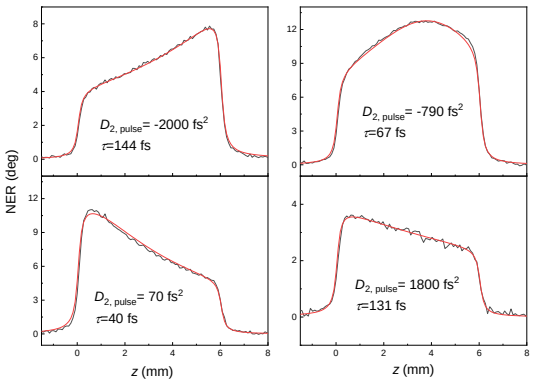

Figure 1: Four NER signals obtained in a 6 mm SF6 sample with pulses at 780 nm with different input chirp ($D_{2,\text{pulse}}$) and fixed τ_0(TL)=40 fs. The black (red) line is the experimental (theoretical, Eq. 1) results.

In our method, we need to know the TL pulse width or the τ_0. For Gaussian pulse, τ_0, can be obtained by measuring the laser pulse bandwidth:

$$\tau_0 = \frac{0.44\lambda^2}{\Delta\lambda c}, \tag{3}$$

where c is the speed of light in vacuum, λ is the central wavelength, and $\Delta\lambda$ is the pulse bandwidth (FWHM) measured by a spectrometer. A good agreement was observed between the experimental and theoretical data (Fig. 1), indicating that the GDD is the dominant dispersion. For example, by fitting the experimental data of Fig. 1, using the correct sample' dispersion ($V_{2,\,SF6}$(780 nm)=206 fs^2/mm) and the nonlinear refraction of SF6, n_2,(23×10^{-20} m^2/W), it was possible to obtain all the beam parameters such as the z_0=0.07 mm, w_0=4.2 μm, I_0~187 GW/cm^2 (for TL pulse), and the dispersion $D_{2,\text{pulse}}$ of the input pulse. Measurement in LaSF-N30 provided the same results. Although this method can provide the absolute value of the material's nonlinearity, it is interesting to use a well-known reference sample to reduce the uncertainty.

Finally, we also did a complete measurement of the width of the pulses delivered by an OPA from 460 nm up to 2700 nm to demonstrate the good bandwidth capability of our method. It is worth pointing out that due to the single-beam configuration and degenerated third-order effect, phase-matching is not a problem. To support our pulse width measurements, we did simultaneous AC measurements using a commercial system (GECO Scanning Autocorrelator from Light Conversion).

In summary, a simple single-beam NER measurement in thick samples was proposed to measure both the pulse properties and n_2, by NER measurements using the sample dispersion as a reference. The pulse width measurements were in good agreement with the ones obtained with a commercial AC device. The simultaneous measurements bring great potential for better materials' third-order nonlinear refraction characterization since the measurements are obtained with the same apparatus and at the same experimental conditions. In addition, our pulse measurements can be a good option when other traditional methods cannot be employed.

References

[1] R. Trebino, Frequency-resolved optical gating: The measurement of ultrashort laser pulses, Kluwer Academic Publishers, Boston (2002).

[2] J. A. C. Gomes, E. C. Barbano, and L. Misoguti, "Cross-section profile of the nonlinear refractive index of Gorilla Glass obtained by nonlinear ellipse rotation measurements," Applied Optics 58, 7858 (2019).

[3] S. De Silvestri, P. Laporta, and O. Svelto, "The role of cavity dispersion in CW mode-locked dye lasers," IEEE J. Quantum Electron. QE-20, 533 (1984).