Dispersion of nonlinear refraction and two-photon absorption using a white-light continuum Z-scan

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Abstract: We use a white-light continuum (WLC) Z-scan technique to measure the degenerate two-photon absorption spectrum and associated dispersion of the nonlinear refraction in ZnSe. The spectral components of the WLC source are separated by using a narrow band variable filter to minimize nondegenerate nonlinearities. We observe a change in sign of the ultrafast nonlinear refractive index around 0.7 of the bandgap energy as predicted by theory.

OCIS codes: (190.0190) Nonlinear Optics, Materials; (300.0300) Spectroscopy, Nonlinear

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1. Introduction

Specific applications require nonlinear properties of materials to be known over a broad wavelength range. The database for the spectral dependence of the nonlinear absorption and nonlinear refraction of materials is sparse. The methods for measuring nonlinearities are often time consuming and prone to errors of interpretation. For example, distinguishing ultrafast nonlinearities such as two-photon absorption, 2PA, from cumulative nonlinearities of, for example, excited-state absorption; or distinguishing bound-electronic nonlinear refraction from free-carrier refraction, can be difficult. Key to our understanding of these nonlinearities in many materials is the development of a large and reliable database of nonlinear spectral properties. For example, the spectral properties are of importance for determining structure-property trends in organic materials, where knowledge of the wavelengths of the peaks of the nonlinearities is important. Here we introduce a method for very rapid and simultaneous determination of both the spectrum of nonlinear absorption (NLA) and dispersion of nonlinear refraction (NLR).

Recently, a WLC was used as an intense source to measure degenerate 2PA over a broad spectral range [1]. We then introduced the "WLC Z-scan" as a method for rapid and broadband characterization of the degenerate NLA and NLR and we demonstrated this method for NLA [2]. At nearly the same time the authors of reference [3] introduced a similar technique that required a separate calibration and ignored nondegenerate 2PA. In our method an intense, broadband, WLC was used to replace the single wavelength source that is conventionally used in a Z-scan experiment. A glass prism and a ZnSe window were employed to spatially and temporally separate the spectral components of the WLC source before entering the nonlinear sample to minimize the nondegenerate 2PA process, which we found was significant in the absence of such separation. To confirm the validity of the method, we measured the degenerate 2PA spectrum of ZnSe. The experimental results showed good agreement with theory [4] and with our previous measurements at specific wavelengths, verifying the validity of our method.

In this paper, we demonstrate that the method can be extended to the "closed aperture" Z-scan for NLR measurements. As in the above 2PA measurements, simultaneous NLR measurements at all wavelengths would also require separation of the wavelengths to minimize nondegenerate nonlinearities. However, spatial separation presents difficulties in using the aperture needed for closed aperture Z-scans. Temporal dispersion of the continuum can broaden the pulses and may significantly reduce the irradiance, thus weakening the nonlinear signal. Here we present an alternative approach for both NLA and NLR, where we separate the WLC wavelengths with a variable frequency bandpass thin-film filter. This "linear variable filter" (LVF) method provides an absolute measurement of the degenerate nonlinearities of the sample under investigation with the high sensitivity of the normal Z-scan technique [5]. To confirm its validity, we test this new procedure on the well-characterized semiconductor, ZnSe [4,6,7,8]. We demonstrate the use of this technique for simultaneous measurement of both NLA and NLR.

2. Experiment: method and results

We used a regeneratively amplified femtosecond laser system which provides laser pulses at 775 nm of 150 fs (FWHM) duration, with an energy of 1 mJ/pulse at a 1 kHz repetition rate.

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For WLC generation, we focused ~10μJ of this beam with an f=15 cm lens into a 3 cm long cell filled with distilled water. A short-pass filter was used to remove the 775 nm pump and the infrared part of the continuum. The spectrum of the WLC after passing through the low-pass filter is shown in Fig. 1.

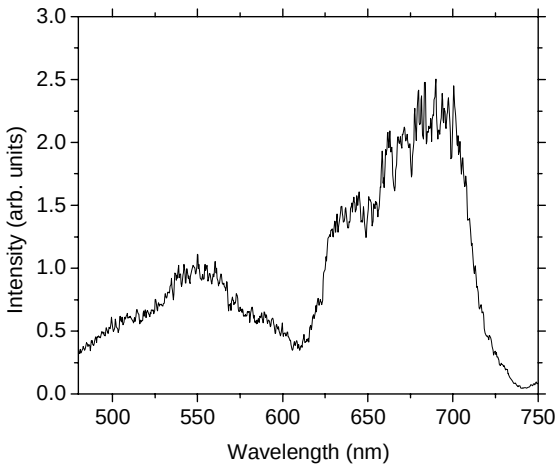

Fig. 1. WLC spectrum after passing through the low-pass filter

To avoid nonlinear processes such as nondegenerate 2PA, cross-phase modulation, etc., we spectrally separate the WLC wavelengths with a narrowband LVF, manufactured by Ocean Optics, Inc.[9]. This acts as a bandpass filter with spatially-varying center wavelength. The bandpass width can also be varied. By focusing the WLC beam through the filter we were able to obtain a transmitted beam having a spectral bandwidth of 12-15 nm (FWHM) over the wavelength range 500 to 710 nm. To obtain Z-scans over this wavelength range, at each Z-position we scan the full wavelength range by translating the LVF through the beam. The sample is translated along Z after each wavelength scan, and the process is repeated until the entire Z-scan is obtained. In practice, we use two computer driven translation stages and, at the 1 kHz laser pulse repetition rate, averaging 500 pulses per data point, a single experiment takes ~8 minutes to give the full spectrum of NLA and full dispersion of NLR. The experimental setup is shown in Fig. 2. After the WLC beam is focused onto the LVF, the single wavelength beam transmitted by the LVF is approximately recollimated, then divided by a beamsplitter into a reference arm and a signal arm. After focusing the beam through the sample, we use a second beamsplitter, to split the transmitted beam into two arms. On one arm, we use a lens to collect all the energy on the detector ("open aperture" Z-scan). On the other arm we place an aperture which transmits only ~40% of the beam ("closed aperture" Z-scan). Nonlinear absorption coefficients are extracted from the transmission curves obtained in the open aperture experimental arm. The experimental curves obtained in the closed aperture arm contain information about both nonlinear absorption and nonlinear refraction. It was shown in reference [6] that, for small signals, the separation of the nonlinear refraction from 2PA can be done by dividing the closed aperture normalized Z-scan by the open aperture one.

To fit the Z-scan transmission curves corresponding to open [6] and closed [10] apertures, we need to know the energy, the beam waist and the pulsewidth for each spectral component. The energies measured were in the range of 0.3 -2 nJ over the range 500 nm - 710 nm. We

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determined the beam waist for different wavelengths transmitted by the LVF by using the knife-edge scanning technique. For pulsewidth measurements we used an Optical Kerr Gate experiment (OKG) [11], where the Kerr medium was a 1mm thick cell filled with CCl_4. From the width of the cross-correlation traces we were able to determine the pulsewidth for different wavelengths transmitted by the LVF. The value determined was ~165 fs FWHM in the range 500 nm – 750 nm.

In order to test this technique we measured the 2PA spectrum and the associated bound electronic n_2 dispersion in ZnSe. We chose ZnSe since we had measured its 2PA and n_2 at several wavelengths previously [4,6,8]. The thickness of the sample was 2.0 mm for which the thin sample approximation for diffraction is valid [6]. However, ZnSe has a large group velocity dispersion (GVD) and we must account for GVD-induced pulsewidth broadening as the pulse passes through the ZnSe sample. To measure the effect of the GVD on the pulsewidth in the ZnSe, we repeated the OKG experiment with the sample placed in the WLC path. This revealed that the broadening was significant, increasing the pulse width from 165 fs FWHM to 260 fs at 710 nm and from 165 fs to 400 fs at 560 nm.

In order to analyze the Z-scan we need to determine the irradiance within the sample. For this, we begin with the experimentally measured pulsewidths without and with the ZnSe in the beam (corresponding respectively to the pulsewidths at the front and at the back of the sample). We then compare the experimental value after the ZnSe sample with that obtained from the following theoretical calculation. Considering the pulse entering the sample to be linearly up-chirped, we can calculate the chirp parameter from its spectral width (determined from the spectrometer) and its pulsewidth (obtained from the OKG measurement). The pulsewidth at the output of the sample is then calculated by taking into account the GVD induced pulse broadening. The experimental pulsewidths were between 8% and 17% longer than theoretical, with an average difference of 14% at all wavelengths. Therefore, to obtain a good value for the average irradiance within the sample, we average the experimentally measured input pulsewidth with the calculated output multiplied by 1.14, i.e. $(\tau_{input}+1.14\tau_{output})/2$. We used the average value of 1.14 because we measured the effect of broadening due to ZnSe for only 4 wavelengths.

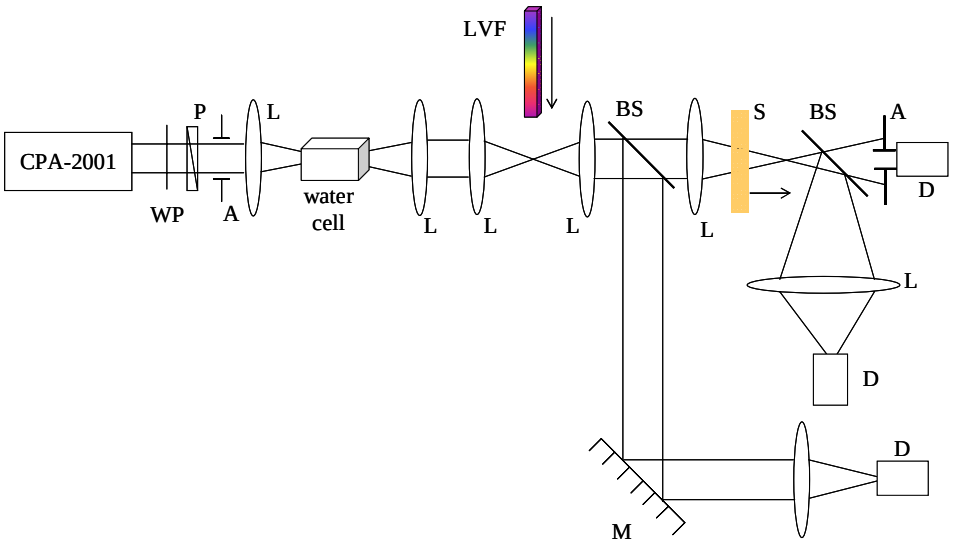

Fig. 2. WLC Z-scan experimental setup. CPA-2001-femtosecond laser source; WP-waveplate; P-polarizer; L-lens; LVF-linear variable filter; M-mirror; BS-beamsplitter; S-sample; A-aperture; D-detector

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The experimental Z-scan curves (open aperture and its corresponding divided closed aperture) are shown in Fig. 3 for two different wavelengths.

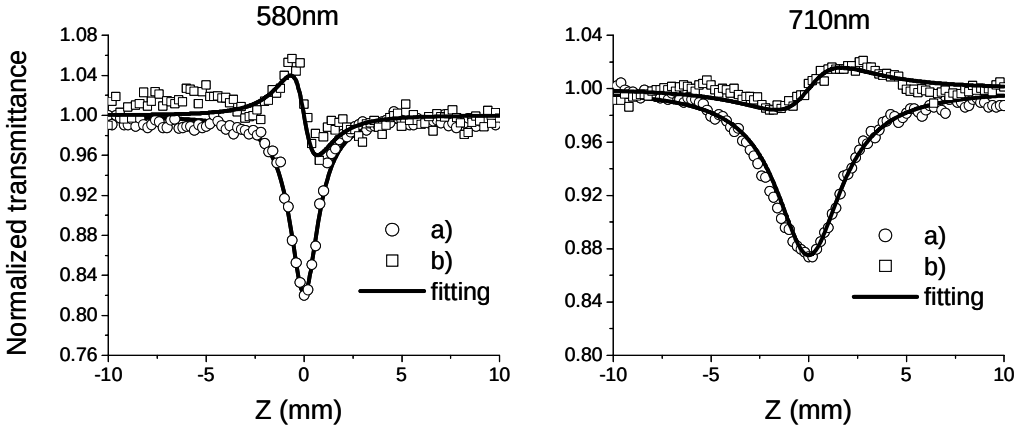

Fig. 3. Z-scan data at 580nm and 710nm a) open aperture and b) closed aperture (the result of the division with open aperture). Here each data point is the average of 500 pulses.

The solid lines represent fits [6,10] to the experimental curves used to extract values of the 2PA coefficient, β (from the open-aperture Z-scan curves) and the values of the nonlinear refractive index, n_2 (from the closed-aperture Z-scan curves). We notice that n_2 changes sign from negative at 580 nm to positive at 710 nm, as predicted by theory in reference [8]. To our knowledge, this is the first time the sign change of n_2 around 0.7 of the energy bandgap has been demonstrated by tuning the wavelength. The values of β and n_2 obtained from fits at different wavelengths are presented in Fig. 4 along with the theoretical predictions of references 4 and 8. The β values have an estimated experimental error of approximately 20%. The errors for n_2 values are between ±30% and ±80%, the larger errors corresponding to the n_2 values close to zero as the transmittance curves were quite noisy.

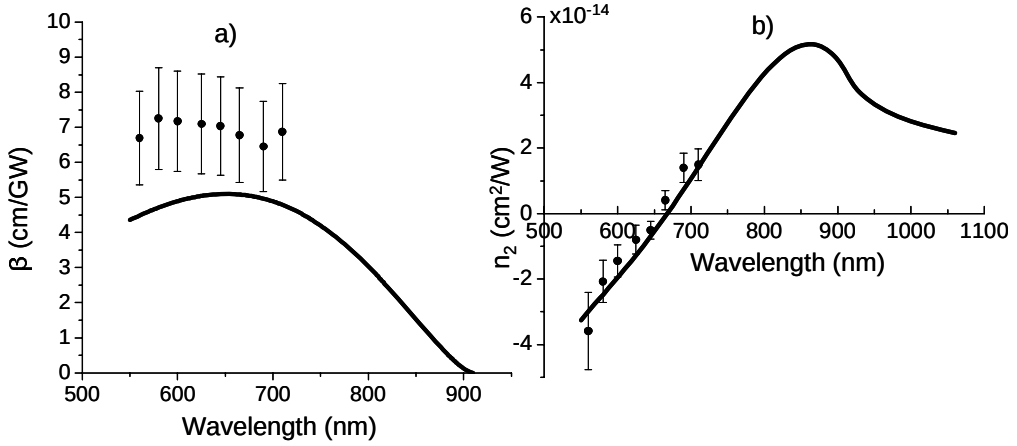

Fig. 4. a) 2PA coefficient and b) nonlinear refractive index coefficient obtained from theory and from the experimental data fittings

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3. Conclusion and future work

We have proposed and demonstrated a sensitive method for rapid and simultaneous characterization of a material's degenerate nonlinear absorption spectrum and the dispersion of its nonlinear refraction. We experimentally verified the method on the well-characterized semiconductor, ZnSe. The good agreement between experimental results and theoretical calculations for the 2PA spectrum and n_2 dispersion attests to the potential of this technique. Extension of this technique to materials with considerably smaller nonlinearities will require higher spectral energy densities for the WLC. High energy WLC sources have already been demonstrated [13], but unfortunately, above a certain threshold, the beam breaks into small filaments making a Z-scan experiment impossible. A promising possibility is to use a WLC generated in hollow fibers which appears to exhibit both high spectral energy densities and good spatial properties [14, 15].

Also, linear variable filters that operate in different spectral regions will allow for measurements over a broader spectrum, thus helping to complete the database of the spectral dependence of the degenerate nonlinear absorption and refraction of materials.

The results of this method when combined with data from our WLC femtosecond pump-probe method [7] yielding the nondegenerate nonlinear absorption with temporal information, give us a very complete picture of the nonlinear absorptive and refractive properties of a material.

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