Single-beam differential z-scan technique

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We report a single-beam, differential z-scan technique with improved sensitivity for the determination of nonlinear absorption and refraction of materials. A sample is scanned in the direction of beam propagation as usual, but, in addition, its longitudinal position is dithered, producing a detector signal proportional to the spatial derivative of only the nonlinear transmission and therefore giving a background-free signal; the nonlinear transmission for any spatial position of the sample can be recovered by simple integration. For both open and closed aperture scans in GaP, we find an improvement in the signal-to-noise ratio of $>5\times$ compared with a balanced z-scan setup, but this can be improved with apparatus optimization. Nonlinear phase distortions $<\lambda/1500$ are obtained with a 78 MHz, mode-locked Ti:sapphire laser. © 2007 Optical Society of America

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1. Introduction

The z scan is a simple technique[1] for determining the absorptive and refractive nonlinear optical properties of matter. In essence, the sample is scanned through the focal point of a high-intensity beam, and the exiting beam intensity and/or spatial profile are measured. The original method suggested the use of a single beam incident on the sample. While, in principle, the method is simple, in practice, since lasers possess intensity fluctuations and the detector measures intensity loss due to both linear and nonlinear processes, the sensitivity is compromised. To overcome some of these limitations, most schemes often employ a reference arm and associated detector. Additional modifications have also been proposed to improve the sensitivity or speed of the technique,[2–9] often at the expense of simplicity.

In this paper, we demonstrate a background-free, single-beam z-scan technique with improved sensitivity. The main innovation is to dither the longitudinal position of the sample as it is being scanned in the beam. By using a short focal length lens with a corresponding short Rayleigh length one can rapidly oscillate the sample over distances comparable to, but less than, the Rayleigh length with a piezotransducer. This oscillatory motion induces a periodic modulation of the beam intensity at the sample, which in turn produces a modulation of the transmitted light and its spatial profile if the sample has a nonlinear response. The modulated component of the detector signal, which can be obtained by lock-in processing, is proportional to the spatial derivative of only the sample's nonlinear transmission and therefore provides a background-free measurement. By simple integration one can recover the sample's nonlinear transmission or, for closed aperture scans, nonlinear refraction. We refer to this as a differential z-scan technique. With a small focused beam waist one can also take advantage of a high-repetition rate, low-noise laser source although this is not essential for the main idea presented here.

Falconieri *et al.*[6] also considered a modulation of the sample's position, but that method can be thought of as a rapid scanning technique to obtain the intensity-dependent transmission of the sample. The (conventional) z-scan traces obtained from each cycle are averaged to increase the signal-to-noise (S/N) ratio. The technique presented here gives the differential transmission as a function of scan position and is not sensitive to total intensity transmission, and, e.g., linear absorption and refraction effects. This differential method therefore has advantages similar to those of other differential techniques or photomodulation type experiments such those used in time-resolved pump–probe experiments.[10] The sensitivity of this differential scheme is demonstrated using a GaP sample in both open and closed aperture z-scan optical configurations. Compared with the well-established bal-

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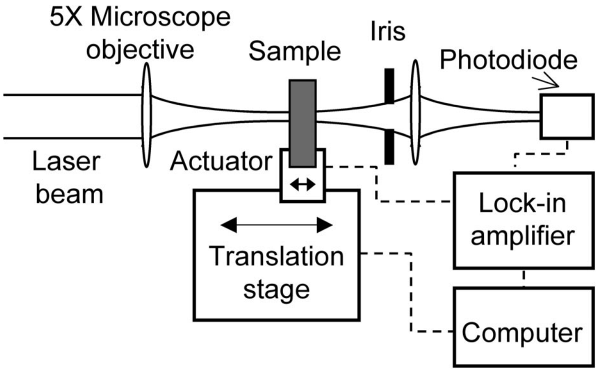

Fig. 1. Differential z-scan experimental setup.

anced z-scan technique, we obtain a >5× increase in S/N ratio without particular attention being paid to configuration optimization. In particular, nonlinear phase distortions as low as $\lambda/1500$ are readily detectable with the present differential z-scan technique.

2. Experimental Technique

The experimental setup for the determination of the differential z scan is shown in Fig. 1. A mode-locked Ti:sapphire laser produces 150 fs pulses at a repetition rate of 78 MHz with a carrier wavelength of 830 nm (1.5 eV). Note that this photon energy is clearly below the bandgap, so no linear absorption is expected. The pulse train is tightly focused by a 5× microscope objective (numerical aperture of 0.12) and passed through the sample, a GaP single-crystal wafer, located near the focal plane. The beam is then refocused with a lens onto a silicon photodiode. As the sample is slowly scanned by a translation stage through the focal point along the beam axis (z) a piezoelectric actuator (Physik Instrumente P-280.10) oscillates the sample at frequency f along z with amplitude L. The amplitude L is chosen to be comparable to the beam's Rayleigh range, $z_0 \sim 60\ \mu$m, yet smaller than the Rayleigh range inside the medium, which has a linear index of refraction of $n = 3.17$. The transmitted intensity around a given position z' can then be written as $T = T(z') + (\partial T/\partial z)_{z'} L \sin 2\pi f t + \ldots$. The amplitude $[(\partial T/\partial z)L]$ of the component at frequency f is detected using a lock-in amplifier with the driving signal of the piezo used as a reference. The use of a lock-in amplifier allows one to suppress noise outside a narrow frequency interval. Note that for $L \lesssim z_0$ only the first derivative of the transmission signal is obtained [the first correction term is proportional to $(\partial^3 T/\partial z^3)$]. Of course, the overall modulated signal amplitude is ultimately limited by the maximum induced change in the transmission dictated by nonlinear refraction or absorption. In all cases, the z-scan signal $[T(z)]$ is obtained by simple numerical integration.

3. Results and Discussion

Figure 2(a) shows an open aperture differential z-scan trace of a 430 μm thick [100]-oriented GaP single-crystal wafer for a pulse energy of only 0.13 nJ. The piezoactuator was operated at $f = 20$ Hz with $L = 45\ \mu$m. The background-free signal has a dispersive shape, changing sign as the sample moves through the focal plane taken to be at $z = 0$. The shape of the differential signal $(\partial T/\partial z)L$ is readily understood if, as is usually the case, $T(z)$ has a Lorentzian shape.[1] For Fig. 2(a), the spatial extent of the differential z-scan trace is larger than z_0 since the GaP wafer is significantly thicker than z_0. Figure 2(b) displays the result of an integration of the modulation signal in Fig. 2(a) after dividing by L. The curve, which shows very low noise, has the shape of a conventional z scan with a maximum $\Delta T/T = 0.08$. The dashed curve in Fig. 2(b) is a fit to the data employing the theoretical expression for a thick medium.[11] For a peak focused laser intensity of $I_0 = 1\ \mathrm{GW/cm^2}$, a two-photon absorption coefficient of 5 cm/GW is obtained, in reasonable agreement with a theoretical value of 1 cm/GW obtained from the two-parabolic-band model[12] of a direct gap semiconductor of $E_{G,\mathrm{direct}} = 2.78$ eV. Note, however, that GaP has also an indirect gap of $E_{G,\mathrm{indirect}} = 2.26$ eV and so the model may not be completely appropriate.

We emphasize that the intrinsic advantage of obtaining differential z-scan data such as in Fig. 2(a) over a conventional z scan is related to the use of a

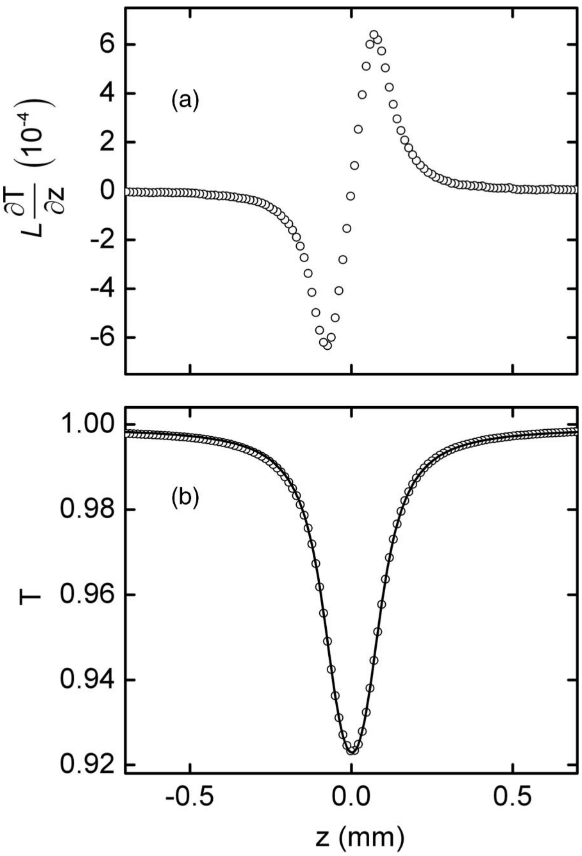

Fig. 2. Open aperture differential z-scan transmission of GaP. (a) Output from lock-in amplifier. (b) Integrated signal (transmission) as a function of the displacement (circles) and numerical fit (curve) based on Ref. 11.

lock-in amplifier locked to the periodic sample motion. The detected response is only sensitive to a signal contained in a narrow frequency range around f. Slow fluctuations of the laser intensity therefore do not compromise the sensitivity of this background-free technique. On the other hand, a conventional z-scan trace would show a variation of the background intensity not related to any nonlinear optical property, thus limiting the sensitivity, especially in the case of small nonlinearities. For the same reason, other major sources of noise such as pointing instabilities of the laser are also drastically reduced in the differential configuration.

We note that the overall amplitude of the differential signal presented in Fig. 2(a) depends on the frequency f applied to the piezoactuator. We find that the oscillation amplitude and thus the observed modulation signal scales approximately as f^{-1}. For our geometry, $f = 20$ Hz offers a good compromise, representing a spatial motion over a significant fraction of the Rayleigh length with an efficient and rapid lock-in analysis. For example, scans over an interval of $20z_0$ can be easily obtained in <1 min. Note that L has to be known in order to convert the differential z-scan data into $T(z)$. The L can be measured easily by comparing both differential and conventional z-scan techniques using a moderately nonlinear material. In our case, the setup was calibrated using both the GaP wafer of this study and a sample of silicon on a sapphire layer. Once the L was determined, it was found to be independent of the sample's mass over a broad range of 4 g.

We have also applied the differential z-scan technique to a closed aperture configuration. An iris allowing 70% of the light intensity in the center of the laser mode is inserted in the experimental setup after the sample. The triangles in Fig. 3 show the result for a z scan of the GaP sample with the differential technique for a laser pulse energy as low as 0.13 nJ. The trace is already integrated along the z axis as discussed above. The z-scan data are divided by the open aperture signal to eliminate two-photon absorption effects. Moreover, traces obtained at very low incident power were subtracted from the data to remove effects of sample inhomogeneity and multiple reflections from the sample surfaces. From a fit to the closed aperture z-scan data, we obtain a nonlinear refractive index of $n_2 = 5 \times 10^{-18}$ m^2/W for [100]-oriented GaP, also in good agreement with the theoretical estimate[12] of 3×10^{-18} m^2/W.

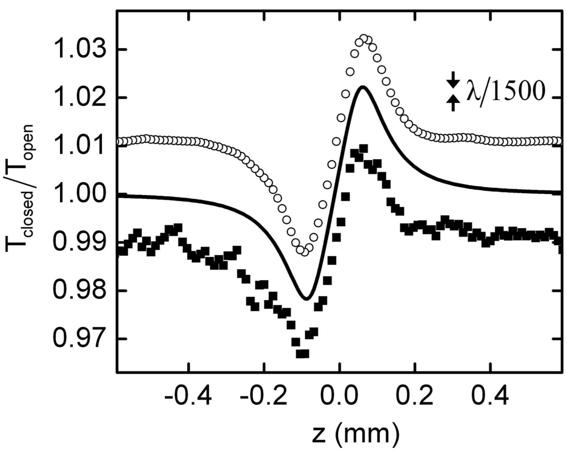

Fig. 3. Normalized transmission for closed aperture integrated differential z scan (circles) and conventional z scan (squares). The two experimental traces are displaced by $+1\%$ and -1% along the y axis for clarity. The numerical fit (curve) is based on Ref. 11.

To be able to compare the sensitivity of the differential method to established techniques, we performed closed aperture z scans using a conventional balanced detection scheme. For these measurements, a reference arm was introduced, detecting the incident laser intensity with a second photodiode.[1] The z-scan data are then extracted by analyzing the difference between the intensity transmitted through the sample and the real time intensity measurement in the reference arm. Again, these data are corrected with results obtained at a very low incident power to eliminate any linear absorption contributions. Results obtained with this balanced technique are displayed in Fig. 3 for the same excitation conditions as for the differential configuration. Comparing the two experimental traces, we find good overall agreement of the two z-scan methods. However, the differential technique offers a superior S/N ratio. From the data in Fig. 3, we estimate that the minimum peak-to-valley transmission difference we can resolve with the closed aperture differential technique is as low as $\Delta T/T = 0.0015$, which corresponds to a sensitivity to a nonlinear phase distortion of $\lambda/1500$. This value is a clear improvement of the previously reported[13] $\lambda/300$ for the conventional z-scan technique. We note that even higher sensitivity can be achieved by using a piezoelectric actuator with larger maximum amplitude. A larger oscillation amplitude does not alter the noise level but increases the signal as noted above.

4. Summary

In conclusion, we have demonstrated a single-beam differential z-scan technique that enhances the sensitivity for determination of nonlinear absorption and refraction. The background-free scheme takes full advantage of a lock-in analysis of the transmitted intensity referenced to a fast sample oscillation along the beam axis. The sensitivity to nonlinear optical phase distortions is as low as $\lambda/1500$. This differential technique can also be combined with more elaborate z-scan setups such as an excite-probe[9,14] configuration to time-resolved optical nonlinearities. No attempt was made to optimize the experimental setup. Other choices of the dither amplitude or the frequency could improve the sensitivity.

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References

1. M. Sheik-Bahae, A. A. Said, T.-H. Wei, D. J. Hagan, E. W. Van Stryland, "Sensitive measurement of optical nonlinearities using a single beam," IEEE J. Quantum Electron. **26,** 760–769 (1990).
2. W. Zhao and P. Palffy-Muhoray, "Z-scan technique using top-hat beams," Appl. Phys. Lett. **63,** 1613–1615 (1993).
3. T. Xia, D. J. Hagan, M. Sheik-Bahae, and E. W. Van Stryland, "Eclipsing z-scan measurement of $\lambda/104$ wave-front distortion," Opt. Lett. **19,** 317–319 (1994).
4. M. Martinelli, S. Bian, J. R. Leite, and R. J. Horowicz, "Sensitivity-enhanced reflection z-scan by oblique incidence of a polarized beam," Appl. Phys. Lett. **72,** 1427–1429 (1998).
5. I. J. Blewett, J. Stokes, A. Tookey, A. K. Kar, and B. S. Wherrett, "Fastscan z-scan system for determining optical non-linearities in semiconductors," Opt. Laser Technol. **29,** 355–358 (1997).
6. M. Falconieri, E. Palange, and H. L. Fragnito, "Achievement of $\lambda/4000$ phase distortion sensitivity in the measurement of optical nonlinearities by using a modulated z-scan technique," J. Opt. A **4,** 404–407 (2002).
7. C. R. Mendoça, L. Misoguti, and S. C. Zilio, "Z-scan measurements with Fourier analysis in ton-doped solids," Appl. Phys. Lett. **71,** 2094–2096 (1997).
8. T. Catunda, J. P. Andreeta, and J. C. Castro, "Differential interferometric technique for the measurement of the nonlinear index of refraction of ruby and $GdAlO_3{:}Cr^{+3}$," Appl. Opt. **25,** 2391–2395 (1986).
9. J. Wang, M. Sheik-Bahae, A. A. Said, D. J. Hagan, and E. W. Van Stryland, "Time-resolved z-scan measurements of optical nonlinearities," J. Opt. Soc. Am. B **11,** 1009–1017 (1994).
10. J. Shah, *Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures* (Springer, 1996).
11. W.-P. Zang, J.-G. Tian, Z.-B. Liu, W.-Y. Zhou, F. Song, and C.-P. Zhang, "Analytic solutions to z-scan characteristics of thick media with nonlinear refraction and nonlinear absorption," J. Opt. Soc. Am. B **21,** 63–66 (2004).
12. M. Sheik-Bahae, D. J. Hagan, and E. W. Van Stryland, "Dispersion and band-gap scaling of the electronic Kerr effect in solids associated with two-photon absorption," Phys. Rev. Lett. **65,** 96–99 (1990).
13. M. Sheik-Bahae, A. A. Said, and E. W. Van Stryland, "High-sensitivity, single-beam N_2 measurements," Opt. Lett. **14,** 955–957 (1989).
14. H. Ma, A. S. L. Gomes, and C. B. de Araujo, "Measurements of nondegenerate optical nonlinearity using a two-color single beam method," Appl. Phys. Lett. **59,** 2666–2668 (1991).