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Novel single beam technique for measuring nonlinear optical absorption of materials by using an electrically focus-tunable lens

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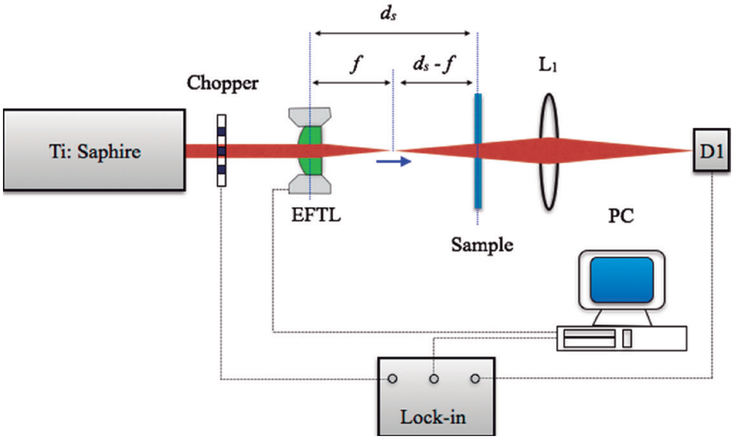

Abstract

We have measured the two-photon absorption coefficients for CdS and ZnSe by using a new single beam setup based on the well-known Z-scan technique. This novel technique, which we have named "f-scan", uses an electrically focus-tunable lens instead of a mechanical translation stage, simplifying the experimental setup and reducing dramatically the cost of equipment while maintaining the experimental sensitivity. Also we found, experimentally, that the technique is more robust to sample surface roughness and multiple scattering because of the elimination of the mechanical motion of the sample. In this paper we present the theory as well as the experimental implementation of the technique. Experimental results for CdS and ZnSe are presented and two-photon absorption coefficients are compared to the well-established values for these semiconductors in the literature. The f-scan opens the door to an affordable nonlinear optical technique for basic research and applications.

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Introduction

After the advent of the laser, several techniques have been used to determine the two-photon absorption (TPA) coefficients of various materials. These techniques were based on measuring the luminescence generated, or the change in the conductivity, or the light transmitted through the material under study (Bechtel & Smith, 1976; Van Stryland, Woodall, Vanherzeele & Soileau, 1985; Van Stryland et al., 1985). In particular, Z-scan (Sheik-Bahae, Said, Wei, Hagan & Van Stryland, 1990), which is based on measuring the change of a focused beam light transmitted through a sample material while it is translated along the optical axis, it has been widely used due to its high sensitivity, relative simple optical setup, and low data processing requirements.

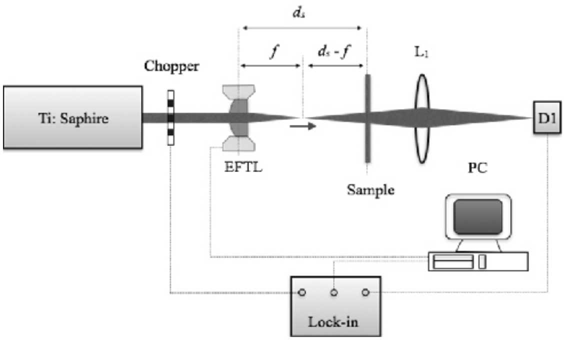
Since Z-scan was proposed, new techniques or modifications to the original idea have been reported, all of them searching for higher sensitivities, simple optical alignments, no displacement of the sample, or cost reductions (Godin, Fromager, Cagniot, Moncorgé & Aït-Ameur, 2011; Gu, Yan, Wang, He & Wang, 2004; Rhee, Byun & Van Stryland, 1996; Tsigaridas, Fakis, Polyzos, Persephonis & Giannetas, 2003), (Xia, Hagan, Sheik-Bahae & Van Stryland, 1994; Yang et al., 2009; Zhao & Palffy-Muhoray, 1993; Boudebs & Cherukulappurath, 2004). For the particular case of TPA measurements, Gu et al. (2004) reported higher sensitivities by using quasi-one-dimensional slit beams, and Yang et al. (2009) introduced phase objects into the measurement system to simplify the optical alignment and avoid sample movement.

Based on a new proposal that tackles the problems of sample movement and complex optical alignments while keeping the high sensitivity and simple data processing requirements of Z-scan (Kolkowski & Samoc, 2014; Serna, Hamad, García & Rueda, 2014), by using an optical setup that implies an important cost-reduction. In the new technique, named "f-scan", the translation stage is replaced by an Electrically Focus-Tunable Lens (EFTL) while the equations are rewritten to consider a varying focal distance instead of a varying sample distance. The new f-scan opens a door to an affordable nonlinear optical technique for basic research and applications. To show the feasibility of f-scan, experimental results for the case of CdS and ZnSe will be presented and will be compared to well established values in the literature (Krauss, 1994; Sutherland, 1996).

Theory and experimental method

The f-scan experimental setup is depicted in Figure 1. A laser Gaussian beam modulated with a chopper impinges on the EFTL, which is a lens that has the capability to vary its focal distance *f* over a specific range when an electric current is applied to it. The EFTL then focuses the Gaussian beam at different positions. The sample is placed at a fixed position d_S inside the range of the EFTL. The light transmitted through the sample is collected by lens L_1 and focused on the photodetector D_1. The output signal of the photodetector is filtered with a Lock-in amplifier and processed with a computer (PC).

Figure 1. *f*-scan open aperture setup



Fuente: by the author.

We measured the transmittance of the nonlinear medium in the far field as a function of the focal distance *f* (see Figure 1). When the distance $|d_S\text{-}f|$ is large the normalized transmittance has a value close to unity because no nonlinear optical effects are produced in the sample. In contrast, small values of $|d_S\text{-}f|$ imply that the laser beam is focused near the sample, thus increasing the optical intensity and generating nonlinear optical phenomena such as nonlinear optical absorption.

For the experimental implementation of f-scan we used a Ti:Sapphire oscillator laser with repetition rate of 90 MHz, pulse width of 30 fs, and laser emission centered at 790 nm. The average output power of the laser was around 300 mW. The power at the entrance surface of the sample was 125 mW due to the use of the optical chopper. The beam diameter at the EFTL was $D=2.1\pm0.1$ mm and was measured by a laser beam profiler. The Electrically Focus-Tunable Lens is an OPTOTUNE-1030 with a tunable focal length between 6.88 cm and 54.71 cm, controlled by an OPTOTUNE lens driver that gives a maximum current of 300 mA with a resolution of 0.1 mA, given a focal length resolution of the order of 0.017 mm. The laser Gaussian beam is focused at quasi-normal incidence in order to eliminate Fabry-Perot effects and multiple reflections. We used a large area Si-photodiode (PDA 50 THORLABS) to measure the transmitted laser light. The current generated by the photodiode is sent to a STANFORD RESEARCH 830 dual channel lock-in amplifier, controlled through a GPIB interface.

The laser beam has a Gaussian spatial profile and a hyperbolic secant temporal profile. Therefore, at the front surface of the sample, the incident beam intensity as a function of focal length f is given by:

$$I_{in}(r,f,t) = I_o(f)\exp\left[-2\left(\frac{r}{w(f)}\right)^2\right]\sec h^2\left(\frac{t}{\tau_0}\right) \qquad (1)$$

In the above equation $\tau_o = \tau / (2ln(1+\sqrt{2}))$ where τ is the full width at half-maximum pulse duration and $I_o\ (f)$ is the peak intensity of the beam as a function of focal length given by:

$$I_0(f) = \frac{2\ln(1+\sqrt{2})P_{ave}}{\tau \cdot v \cdot \pi w_o^2(f)} \qquad (2)$$

Here P_{ave} is the average power of the incident laser beam at the sample, v is the laser pulse repetition rate, and $w_0(f)$ is the beam waist radius as a function of the lens focal length. The beam radius, $w(f)$, at the front surface of the sample is given by:

$$w(f) = w_o(f)\sqrt{1+\left(\frac{d_s - f}{z_o(f)}\right)^2} \qquad (3)$$

Where $z_o\ (f) = (\pi w_o^2\,(f))/\lambda$ is the Rayleigh range and λ is the laser wavelength in free space.

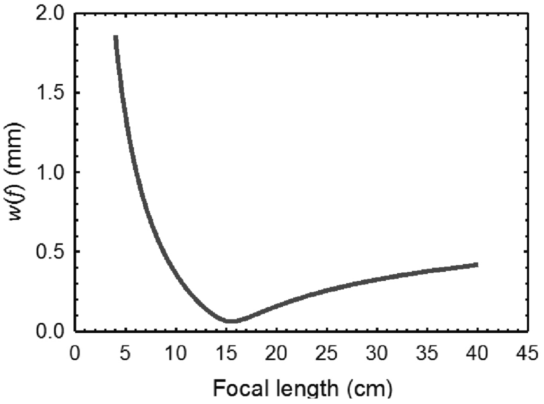
In order to determine the beam waist radius as a function of current and focal length, the lens was characterized with a laser beam profiler. We found that $w_o\ (f)$ can be expressed as:

$$w_O(f) = \frac{2\lambda f}{\pi D}C_f \qquad (4)$$

where $C_f=1.65$ is an EFTL correction factor, D is the diameter of the beam at the lens surface.

Figure 2 shows the dependence of the beam radius at the sample location as a function of the EFTL focal length. Notice the asymmetry relative to the sample location. For f values smaller than ds the radius at the sample increases faster than those for f values larger than d_s.

Figure 2. Typical behavior of beam waist at sample as a function of focal length. This plot was produced using D=2.1 mm and d_s=15.7 mm



Source: by the author.

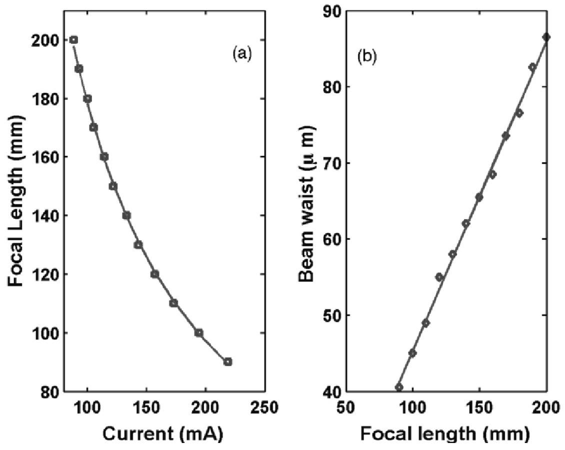
This will cause the shape of the open aperture f-scan to be asymmetric around d_s. Therefore, experimentally, we must normalize the transmitted intensity relative to the intensity corresponding to the shortest focal length used in the experiment.

Experimentally, we found that the focal length as a function of current J can be approximated by the expression:

$$f = \frac{1}{\xi \cdot J + \eta} \qquad (5)$$

Here, $\xi=4.59\times10^{-5}\ mm^{-1}\cdot mA^{-1}$ and $\eta=1.03\times10^{-3}\ mm^{-1}$ are parameters of the EFTL used to convert current to focal length. In Eq. 5 the current is measured in mA . The experimental results obtained with Eq. 4 and 5 are shown in Fig. 3, where it is possible to observe the dependence of the focal length to the current [Figure 3(a)] and the radius beam waist to the focal length [Figure 3(b)].

Figure 3. (a) Focal length as a function of applied current to the EFTL. The points corresponding to experimental values, and solid line to the fitting curve obtained with Eq. 5. (b) Beam waist as a function of focal length, solid points are the experimental values obtained with a CCD camera and the solid line corresponds to Eq. 4



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The intensity of the beam just outside the exit surface of the sample is given by:

$$I_{out}\left(r,f,t\right)=\frac{\left(1-R\right)^{2}I_{in}\left(r,f,t\right)e^{-aL}}{1+\beta\left(1-R\right)I_{in}\left(r,f,t\right)L_{eff}} \qquad (6)$$

where L is the thickness of the sample, R is the reflection coefficient of the sample, α is the linear absorption coefficient, β is the two-photon absorption coefficient (TPA), and $L_{eff}= (1-e^{\alpha L}) / \alpha$ is the effective sample thickness. α for both samples that we have used is less than $0.00005\ cm^{-1}$. The normalized transmission at the detector can be expressed as:

$$T\left(f\right)=\frac{1}{B\left(f\right)}\int_{0}^{\infty} ln\left(1+\frac{B\left(f\right)}{cosh^{2}\left(\rho\right)}\right)d\rho \qquad (7)$$

where $B(f)=\beta(1-R)\ I_0\ (f)L$_effand ρ is an integration variable expressed as ρ=2"ln" (1+√2)t/τ.

Experimental results

Figure 4 and Figure 5 show the experimental results for CdS and ZnSe using the open aperture f-scan technique. We have chosen these semiconductors because their two-photon absorption coefficients are well-known. We used Eq. 7 to fit the experimental data with β as the only fitting parameter. As can be seen the fit is excellent using values of β that are consistent with what is reported in the literature. In the open aperture f-scan data the width of the dip is controlled by the diameter of the beam at the EFTL and the location of the sample from the EFTL. The larger the diameter of the beam the smaller the width of the data profile and as d_s increases the width of the data profile increase. The asymmetry in the data is due to the asymmetry in the radius of the beam at the sample as was mentioned above.

Figure 4. Experimental result (solid points) and fitting (solid line) for CdS

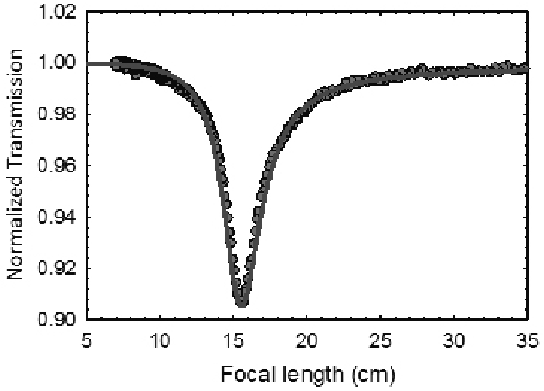

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Figure 5. Experimental result (solid points) and fitting (solid line) for ZnSe

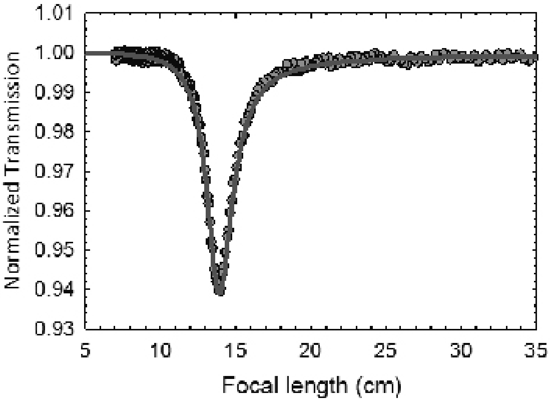

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Table 1 shows some of the parameters used to determine the two-photon absorption coefficients. It is possible to observe that the results are in good agreement with the reported values in the literature (Krauss, 1994; Sutherland, 1996).

Table 1. Physical parameters for the experiment and two-photon absorption

Coefficients

Material	L(mm)	R %	d_s (mm)	β (cm/GW)
Cds	1.0	17	150.7	5.9
ZnSe	0.5	19	140.0	4.9

Source: by the author.

Conclusions

In conclusion, we have analyzed theoretically and performed experimentally a simple technique to measure the nonlinear optical absorption β by using an electrically tunable-focus lens (EFTL). This technique opens the door to a series of practical applications where mechanical translation stages are critical in implementing nonlinear optical techniques. We think that this technique can reduce the complexity of some experimental setups in nonlinear optics and at the same time it reduces the cost of implementation and increases the availability of optical nonlinear laboratories in developing countries where budgetary constraints can be a limiting factor.

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