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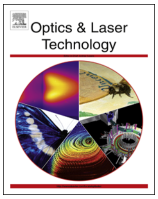

Modified Z-scan set-up using CCD for measurement of optical nonlinearity in PLD carbon thin film

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ABSTRACT

A modified Z-scan set-up utilizing CCD is reported which can be used for open as well as closed aperture configuration in a single scan. The technique is based on recording the transmitted laser beam through the sample on CCD directly. It has the advantage of using the same recorded image for open as well as closed Z-scan for each 'z' position, thus minimizing the effect due to beam pointing instability and background vibrations.

1. Introduction

In 1989, Sheik-bahae et al. reported the Z-scan technique, for measuring the sign and magnitude of the third-order nonlinear refraction coefficient of materials [1]. Ever since, the Z-scan has been widely used technique for measuring the nonlinear optical properties of materials because of its experimental simplicity and sensitivity compared to that of other techniques involving relatively complex experimental setup [2–4]. In this technique, the sample is translated along the optic axis of a focused Gaussian laser beam. The translation of the sample changes the irradiance within the sample, resulting in inducing the modification of intensity dependent optical properties. The intensity transmitted through the sample is recorded as a function of sample position 'z' with respect to the focal plane. The plot of transmitted intensity as a function of 'z' gives the information about the order of the nonlinearity as well as its sign and magnitude. The open aperture (OA) Z-scan determines nonlinear absorption (NLA) coefficient. The closed aperture (CA) Z-scan with appropriate aperture size reflects nonlinear refractive index (NLR) coefficient.

In the present paper, the photodiode in the conventional Z-scan experimental setup was replaced by charge-coupled device (CCD) camera for the measurement of transmitted intensity. The Z-scan measurements using CCD reported earlier were based on (a) subtracting the reference beam from signal beam [5], (b) replacing aperture with an opaque disk [6] and (c) measuring the beam dimension [7]. In the present set-up, the hard aperture was replaced by a software aperture and the intensity was obtained by integrating the image gray values. This modified Z-scan setup using CCD offers several advantages over conventional system using a photodiode: (a) the dynamic range of a CCD detector is very large and its pixel size is in few micrometers, which enhances the sensitivity compared to that of the conventional Z-scan, (b) a suitable synthetic aperture was applied on to the image using Matlab programming rather than the physical aperture, making it independent of the beam pointing instability of the laser, (c) a hard aperture (circular pinhole in conventional Z-scan) does not follow the shape of the beam and so it cannot be used for other than Gaussian beam, whereas the synthetic aperture employed numerically by the Matlab programming, can be easily defined to match the incident laser beam profile, (d) it reduces the experiment running time, as data was obtained for both, open as well as closed aperture configuration in a single Z-scan and (e) focusing lens in front of the detector is not required. In CA Z-scan measurement, an aperture is used to separate out the contribution of absorptive nonlinearity from the nonlinearity due to refractive index. A closed Z-scan curve having equal magnitude of the height of peak and depth of valley is devoid of contribution of NLA [1]. If the aperture size is not appropriate then the CA curve is not symmetric and hence the calculated NLR coefficient will be incorrect. In CA Z-scan, the setup using photodiode, experiment is to be repeated for various apertures to obtain the optimum aperture size such that the peak and valley of the curve are equal. The CCD camera records the profile of the transmitted beam and so the data for the variant aperture size from the same set of images can be obtained by applying the aperture via software after recording the

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full images. Thus the data is extracted for both, OA and CA from same set of images and the effect of any fluctuations in the laser beam is automatically canceled out and simultaneous recording of the reference beam is not required.

2. Experimental details

To demonstrate the modified Z-scan experimental setup, an amorphous carbon thin film was used as a nonlinear medium. The carbon thin film was deposited by the pulsed laser deposition (PLD) technique. The deposition was carried out using a Q-switched Nd:YAG laser (QUANTA SYSTEM HYL-101, 532 nm, 10 Hz) at a base pressure of the order of ~10^{-5} mbar. The film was deposited for 5 min onto fused silica at a substrate temperature of 750 °C using graphite target. The thickness, L, of the film was measured using stylus profilometer (Veeco Dektak 150) and found to be ~30 nm which is much less than the Rayleigh length of the He–Ne laser beam and hence fulfills the thin sample approximation for Z-scan technique. The linear absorption coefficient 'α' was calculated from the absorption spectra using the expression $\alpha=(1/L)\ln(I/I_0)$, where L is the thickness of the film, I is the transmitted intensity through the carbon thin film and I_0 is the incident intensity onto it. The linear absorption coefficient of the carbon film at 632.8 nm was found to be 2.74×10^5 cm^{-1}. Fig. 1 shows the schematic of the modified Z-scan set-up for the measurement of NLA and NLR coefficients. A He–Ne laser (32 mW, MELLES GRIOT 05-LHP-927, 632.8 nm) was focused by a convex lens of focal length of 5 cm. The PLD thin film of carbon under investigation was placed after the lens as shown in Fig. 1. The intensity distribution of the transmitted beam was recorded on a CCD detector (PCO PixelFly) kept at a distance of ~25 cm from the focusing lens. A neutral density (ND) filter was placed in front of CCD to avoid its saturation. An iris diaphragm of the aperture size 6 mm was placed before the ND filter (20 cm from the focusing lens), as shown in Fig. 1, to suppress the scattered light entering into the CCD.

The images of the transmitted beam were recorded by scanning the film 2.0 cm either side of the focal position of the lens. The transmitted intensity through the thin film was obtained by integrating the gray values of the recorded images. The same images were used to obtain the data for CA Z-scan by applying a suitable synthetic aperture the by Matlab program. The aperture size was varied and the ratio of integrated intensity of the masked image (with the synthetic aperture) to that of the entire image of laser beam (S) after passing through the thin film in the range of 0.20–0.60 was measured to determine the phase distortion so as to obtain the optimum value of S.

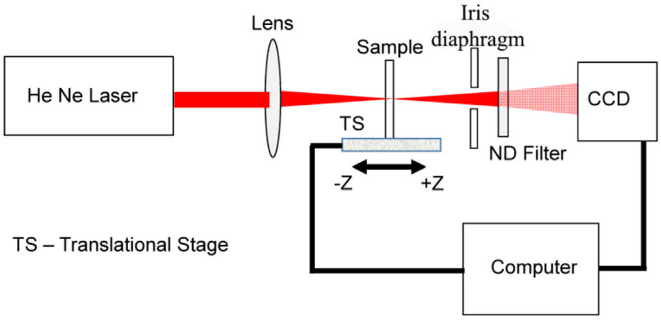

Fig. 1. Schematic diagram of Z-scan setup.

3. Results and discussion

The structural characterization of the PLD carbon thin film was studied by recording Raman spectrum (LabRam HR 800) using He–Ne laser (632.8 nm) as an excitation source. The Raman spectrum of the PLD carbon thin film is shown in Fig. 2. The de-convoluted spectrum shows five peaks; 1604 cm^{-1}, 1586 cm^{-1}, 1347 cm^{-1}, 1326 cm^{-1} and 1104 cm^{-1}. The peak at 1586 cm^{-1} designated as G-band originates from lattice vibrations due to E_{2g} symmetry and corresponds to sp^2 bonding [8]. The peak at 1347 cm^{-1} known as D-band appears due to the presence of structural disorder and corresponds to the breathing modes of A_{1g} symmetry [8]. Depending upon the disorder, the position and width of D band may vary. In the present spectrum, a small peak around 1326 cm^{-1} appeared as a sub-band of D band. The Raman peak towards higher frequency at 1604 cm^{-1} is due to microcrystalline graphite [9]. The origin of peak at 1104 cm^{-1} is not known [10,11]. The presence of sp^2 bonding in the present carbon film furnishes delocalized π-electrons, thus exhibiting the optical nonlinearity.

The recorded CCD image of the transmitted beam through the thin film positioned at 20 mm from the focal point is shown in Fig. 3(a) and (b) for OA and CA Z-scan configuration (for $S\sim0.40$) respectively.

The transmitted intensity for OA was normalized with that of the far field (beyond the Rayleigh length where nonlinear effect does not exist) intensity in order to obtain the normalized transmittance. Fig. 4 shows the normalized transmittance plot as a function of 'z'. The experimental data was fitted to Eq. (1) and is shown as solid line in Fig. 4 [12].

$$T_{op}=1-\frac{c}{1+bz^2} \tag{1}$$

where, $c=\beta I_0 L_{eff}/2^{3/2}$, $b=1/z_0^2$, z_0 is the Rayleigh length, I_0 is the intensity of the laser beam at the focus, β is the NLA coefficient and L_{eff} is the effective thickness of the carbon film which is given by Eq. (2).

$$L_{eff}=\frac{1-\exp(-\alpha L)}{\alpha} \tag{2}$$

The OA Z-scan profile shows valley around the focal position and symmetric to either side of it, indicating the reverse saturation absorption (RSA) effect in the film. The NLA coefficient was calculated from Eq. (1) and found to be 8.22 ± 0.91 cm/W.

In order to determine the NLR coefficient from Z-scan images, the contribution of NLA is to be subtracted by obtaining the closed Z-scan images at optimum aperture size such that the CA Z-scan

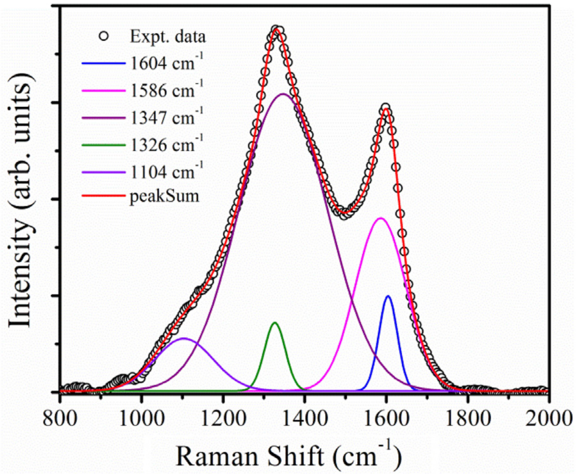

Fig. 2. Raman spectrum of PLD deposited carbon thin film.