PHYSICAL REVIEW A VOLUME 56, NUMBER 2 AUGUST 1997

Theory of *Z*-scan measurements using Gaussian-Bessel beams

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(Received 25 February 1997)

We report a theoretical study of the *Z*-scan technique using Gaussian-Bessel beams. First we find that even the slightest deviation from a perfect Gaussian beam can change the sensitivity of the measurements significantly. Second we demonstrate that the propagation of Gaussian beams modulated by a small Bessel profile can increase the *Z*-scan sensitivity—obtained using conventional Gaussian beams—by a factor of 40 and more. [S1050-2947(97)50708-6]

PACS number(s): 42.65.An, 42.25.Bs, 42.65.Hw

Optoelectronic materials that exhibit a nonlinear refractive index are currently an active area of research partially because of their applications to high-speed all-optical switching. Suitable candidates for all-optical switches, in addition to yielding little absorption, must offer a large and fast response to the inducing pulse. Consequently, one can envision that, by changing the refractive index of a material, the nonlinearities will modify either the direction of propagation, phase, or transmission of another optical pulse through the device.

Since the various intrinsic and extrinsic properties of the material obviously define its possible applications, it has become necessary to define figures of merit for an optical switch—depending on which combination of parameters one wishes to optimize. For this reason it is imperative to make an accurate determination of the material parameters, such as the nonlinear refractive index contributions. Depending on the time scale of the nonlinearity, and on the material, the physical processes that give rise to a photoinduced refractive index change may include *inter alia* virtual electronic processes, free carrier effects, and thermal induced contributions.

Several experimental techniques have been developed over the years to determine the refractive dynamics in optoelectronic materials. One of the most successful experimental methods, originated by Hill *et al.* [1] and quantitatively developed by Van Stryland and co-workers [2,3], has been the sensitive *Z*-scan technique. The *Z*-scan technique exploits the fact that a spatially Gaussian beam can induce spatially dependent refractive index changes, thereby generating an effective lens that causes the beam to focus or defocus [4]. The essence of this technique involves tightly focusing a single Gaussian laser beam into a thin sample. The nonlinear sample consequently imitates a lens and transmittance changes are observed in the far field as the sample is moved through the focal plane; this results in a characteristic peak and valley as the sample is scanned through the beam waist. The far-field diffraction patterns can be subsequently modeled—by employing Gaussian beam optics—to allow the extraction of, for example, the nonlinear refractive index n_2; common theoretical techniques typically apply the Gaussian decomposition method developed by Weaire *et al.* [5]. Although the *Z*-scan technique spatially and temporally averages over effects in the beam, it nevertheless gives quick and useful estimates of the magnitude and sign of the refractive index change.

Recently, improvements and refinements of the *Z*-scan technique have been made, including the ecliptic *Z*-scan (EZ-scan) technique [6], which improves the *Z*-scan sensitivity in the post sample optics by recognizing that the wings of the beam suffer the largest relative irradiance change in an induced lens; a top-hat beam *Z*-scan technique, which increases the sensitivity of the peak to valley in the *Z*-scan trace by a factor of 2.5, compared with that of a Gaussian beam [7]; the two-color *Z*-scan [2], for the study of nondegenerate nonlinearities; the time-resolved *Z* scan [8]; and optically thick *Z*-scan measurements [9–11].

Previously, we reported a very efficient fast-Fourier-transform algorithm to model quantitatively the effects of nonlinear refraction in both ''optically'' thin and thick media; in that work it was demonstrated that sample thicknesses can have very strong effects on the output transmittance [9]. Thus by accounting for internal and far-field nonlinear refractive effects, the method allows one the opportunity to increase experimental measurement sensitivity by scanning thicker samples, and the prediction of, for example, optical switching under more likely practical conditions. In this Rapid Communication, we are not concerned with the influence of internal propagation effects in thick media, but more importantly we are interested in what role the transverse profile of the incident beam plays on the far-field diffraction patterns. Therefore we may employ the thin-sample approximation, and a single discrete fast Fourier transform can be used in the modeling for any arbitrarily shaped input beam [9].

Several years ago, new types of coherent light beams—''diffraction-free beams''—were predicted theoretically [12] and demonstrated experimentally [13]; these somewhat peculiar beams have transverse distributions of the form of a Bessel function of the first kind and zero order, and practically they can be obtained by a simple experimental arrangement involving diffraction from a circular slit. Propagation studies have also been performed for Gaussian-Bessel (GB) beams that can be realized experimentally [14] and also super-Gaussian-Bessel beams [15]. In the present work we focus our attention on the GB beam, because in principle it is easier to obtain experimentally, and an analytical solution of the paraxial wave equation exists for the propagating beam. Propagation of such non-Gaussian shaped beams is important from both the theoretical and the experimental viewpoints. Assuming a GB beam traveling in the $+z$ direction, we can write E as follows [14]:

$$E(z,r,t)=E_0(t)\frac{\omega_0}{\omega(z)}\exp\{i[(k-\zeta^2/2k)z-\Phi(z)]\}$$
$$\times J_0[\zeta r/(1+iz/L)]\exp\{[-1/\omega^2(z)+ik/2R(z)]$$
$$\times(r^2+\zeta^2 z^2/k^2)\}, \quad (1)$$

where $L=k\omega_0^2/2$ is the Gaussian beam diffraction length; $\Phi(z)=\arctan(z/L)$ is the z-dependent phase; $R(z)=z+L^2/z$ is the radius of curvature; and $\omega(z)=\omega_0[1+(z/L)^2]^{1/2}$, where ω_0 is the focal spot size (half width e^{-2} maximum irradiance) of a Gaussian beam. These parameters are identical to those that occur in the description of a Gaussian beam, but the present profile is modified by ζ, where ζ^{-1} is the central maximum width of the Bessel beam; and as expected, for $\zeta=0$, the profile reduces to that of an ordinary Gaussian beam.

First we would like to investigate the sensitivity of ΔT_{PV}, which is the difference between a peak and valley transmittance in a normalized Z scan, when the input beam differs just slightly from that of a Gaussian profile. This is a useful study since often the lasers used in the laboratory do not produce perfect Gaussian beams. For our Z-scan simulations we choose laser pulses that closely resemble those obtained from a frequency-doubled, passively mode-locked, neodymium-doped yitrium aluminum garnet (Nd:YAG) laser, focused to a certain spot size. The temporal shape of the pulse is taken to be of Gaussian profile, with a 36-ps FWHM (full width at half maximum) irradiance. As a nonlinear material we choose the bulk semiconductor ZnSe excited by radiation below the band gap [2]. With an input wavelength of 532 nm, ZnSe is a two-photon absorber, and a cubic refractive nonlinearity can be assumed $(n=n_0+n_2|E|^2/2=n_0+\gamma I)$. The nonlinear parameters were taken to be $\gamma=-6.8\times10^{-14}$ cm^2 W^{-1} ($n_2=-4.4\times10^{-11}$ esu), and for the two-photon absorption coefficient, $\beta=5.8$ cm GW^{-1} [2]. The sample length was chosen to be 2 mm, and the sample-detector distance was 50 cm.

Figure 1(a) depicts a perfect Gaussian beam (solid line) and a modified GB beam with β=0.1 μm^{-1} (dotted line); the FWHM for both beams was chosen to be 19 μm at the focal plane. For the pulses we take an input energy of 0.035μJ, which corresponds to a peak irradiance of 0.21 and 0.22 GW cm^{-2}, respectively, for the Gaussian and GB beams. Transmittance traces were modeled first for an open detector aperture and second with a closed aperture that let in 16% and 7% of the low-intensity light for the Gaussian and GB beams, respectively. For the above parameters, Fig. 1(b) depicts the division of the closed and open results, resulting in a peak before the focal plane and a valley after the focus; this is the case for a negative nonlinear refractive index; i.e., the sample functions as a diverging lens. This situation is reversed for a self-focusing nonlinearity. Note that the Z-scan traces are not completely symmetric because of the role of nonlinear absorption in this regime [2]. The sensitivity of the open Z-scan simulations (not shown) was approximately the same for both beams.

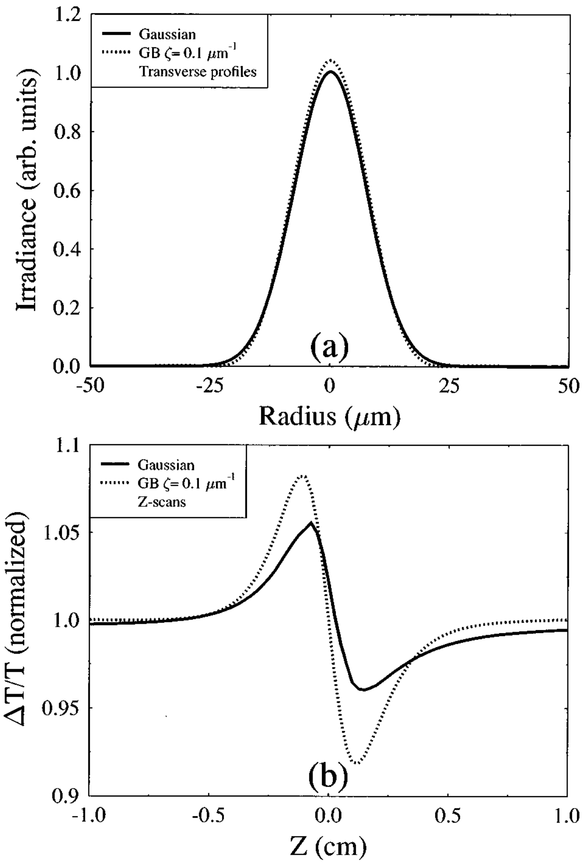

FIG. 1. (a) Transverse irradiance distribution (at the beam waist) of a Gaussian-Bessel beam (ζ=0.1 μm^{-1}) and a Gaussian beam. The FWHM for both beams was 19 μm. (b) Normalized Z scans simulated with an energy of 0.035 μJ, for the beam profiles shown in (a).

As can be recognized from the figure, the ΔT_{PV} of the GB beam is almost double that of the Gaussian beam value, even though their input profiles appear to be very similar. Thus we conclude that one has to be very careful in employing a theory which assumes perfect Gaussian beams, since even for a slight modification of the Gaussian profile, this may lead to erroneously obtained nonlinear coefficients. For example, if we had attempted to reproduce the GB Z-scan trace (assuming an input Gaussian beam), with γ used as a free parameter, we would have then obtained a 30% greater nonlinear refractive index. Therefore before one can reliably employ a Gaussian beam theory for modeling Z-scan measurements, one should take great care in obtaining, experimentally, a very well defined Gaussian spatial profile.

Next we investigate the Z scan so obtained by using a different ζ value of 0.11 μm^{-1} and again compare it to a perfect Gaussian beam with the same FWHM (obtained from the central peak) of 19 μm at the focal plane. These spatial distributions are depicted in Fig. 2(a). As above, the input energy was 0.035 μJ, which corresponds to a peak irradiance of 0.21 and 0.19 GW cm^{-2}, respectively, for the Gaussian and GB beams. Figure 2(b) shows the corresponding

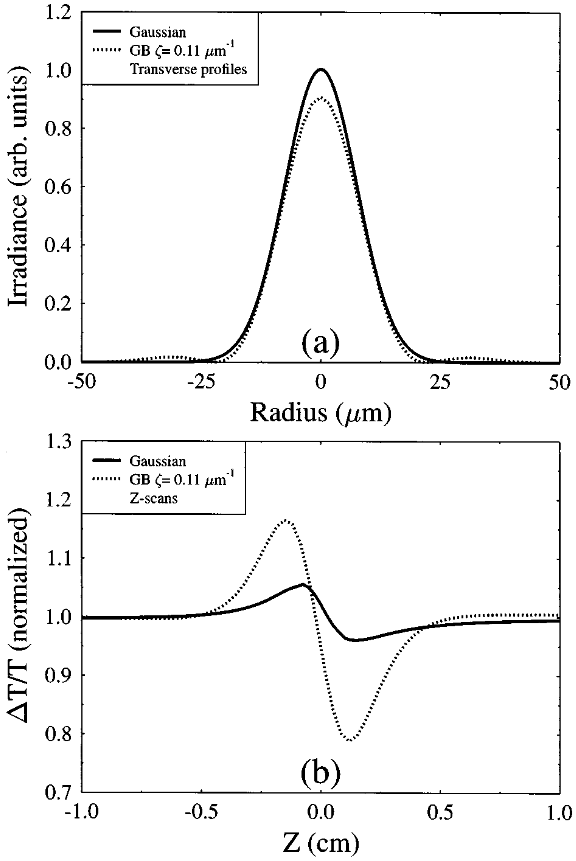

FIG. 2. (a) Transverse irradiance distribution (at the beam waist) of a Gaussian-Bessel beam (ζ=0.11 μm^{-1}) and a Gaussian beam. The FWHM for both beams was 19 μm. (b) Normalized Z scans simulated with an energy of 0.035 μJ, for the beam profiles shown in (a).

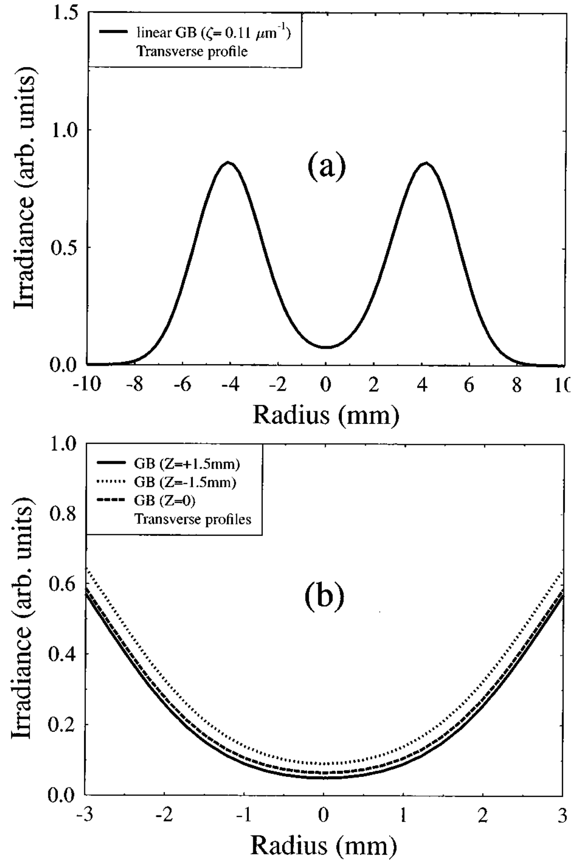

FIG. 3. (a) Transverse far-field irradiance distribution (at the detector plane) of a linear Gaussian-Bessel beam (ζ=0.11 μm^{-1}, FWHM 19 μm). (b) Corresponding nonlinear profiles (at the detector plane) for the sample placed at the focal plane (dashed line), +1.5 cm (solid line), and at −1.5 cm (dotted line).

Z-scan trace for the two beams, and, as can be clearly seen, there is a large enhancement of ΔT_{PV} by about a factor of 4. From these findings, the potential uses of employing GB beams for conducting highly efficient refractive Z-scan measurements are made clear.

This Z-scan technique offers a substantial improvement over conventional Z-scan measurements by exploiting the fact that, in the far field, the center of the GB beam suffers the largest relative irradiance change in an induced lens. To highlight this scenario, we depict in Fig. 3(a) the resulting far-field GB transverse profile at the detector plane for a linear input beam [Fig. 2(a)], where the qualitative features are in agreement with the results shown in Ref [14]: the strong interference is manifested in a characteristic diffractive spreading associated with GB wave propagation. The corresponding nonlinear transverse profiles falling on the detector are shown in Fig. 3(b), where the chosen aperture was from negative to plus 0.18 mm; the dashed line corresponds to the case where the sample is placed at the detector plane, while the dotted and solid lines correspond to prefocal and postfocal Z scans at $\pm$1.5 cm. For comparison, the Gaussian results are shown in Figs. 4(a) and 4(b); undisputably, as a consequence of the unique propagation behavior of the Gaussian-Bessel beam, the relative changes for the GB beam on the detector are much larger.

Finally we investigate the Z scan obtained by using a larger ζ value of 0.12 μm^{-1}, and again compare it to a perfect Gaussian beam with the same FWHM (obtained from the central peak) of 19 μm at the focal plane. These spatial distributions are depicted in Fig. 5(a). As above, the input energy was 0.035 μJ, which corresponds to a peak irradiance of 0.21 and 0.16 GW cm^{-2}, respectively, for the Gaussian and GB beams. Figure 5(b) shows the corresponding Z-scan trace for the two beams, and as can be clearly seen there is a huge enhancement of ΔT_{PV} by almost a factor of 40; note that the Gaussian Z scan has been increased by a factor of 10 for clarity. As a matter of interest, the sensitivity of the GB beam open-aperture Z scan (not shown) was approximately 40% less than that of the Gaussian beam Z scan, which is as expected because of the reduced input peak irradiance.

We would like to mention that, although the EZ scan [6] also offers an increase by an order of magnitude of ΔT_{PV}, this is achieved by rearranging the postsample optics, where the far-field aperture is replaced by a 99% obstruction disk that blocks most of the light. In the present study we could also increase the sensitivity ever further, by adjusting the postsample optics, and for this first study our parameters have in no way been optimized yet. However, for the GB beam propagation, as in the EZ scan, there is a limit to the maximum sensitivity that can be obtained, since the energy reaching the detector becomes too small to detect. Neverthe-

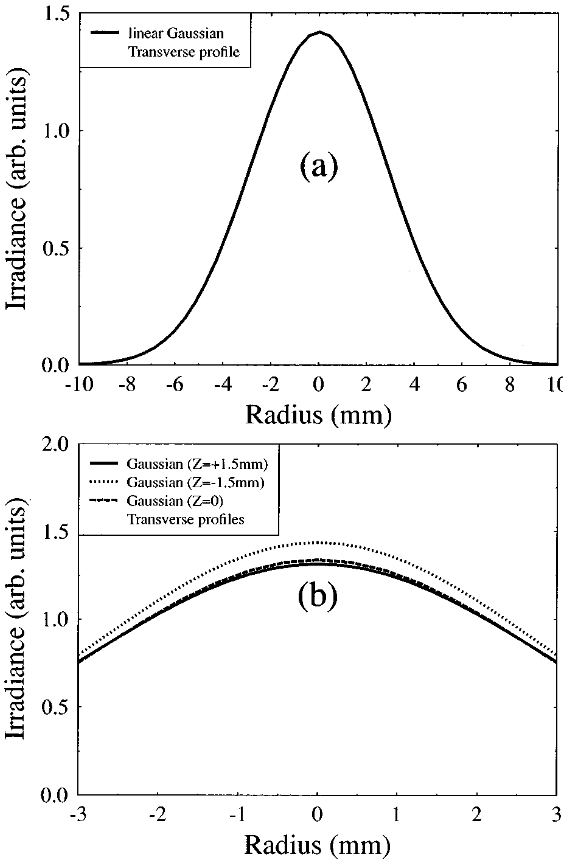

FIG. 4. (a) Transverse far-field irradiance distribution (at the detector plane) of a linear Gaussian beam (FWHM 19 μm). (b) Corresponding nonlinear profiles (at the detector plane) for the sample placed at the focal plane (dashed line), $+1.5$ cm (solid line), and at -1.5 cm (dotted line).

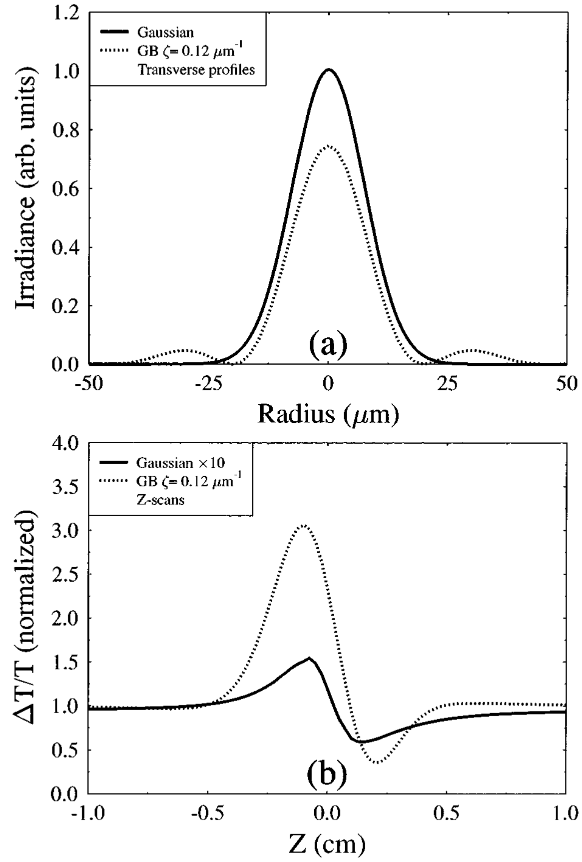

FIG. 5. (a) Transverse irradiance distribution (at the beam waist) of a Gaussian-Bessel beam ($\zeta = 0.12\ \mu\mathrm{m}^{-1}$) and a Gaussian beam. The FWHM for both beams was 19 μm. (b) Normalized Z scans simulated with an energy of 0.035 μJ for the beam profiles shown in (a). For clarity the Gaussian Z scan has been increased by a factor of 10.

less, the fact still remains that very large sensitivity enhancements should still be obtainable, without adjusting the delicate postsample optics, by employing GB beams.

In summary, the characteristic peak to valley (ΔT_{PV}) obtained by employing a Z-scan technique was found to differ considerably by using input spatial beams that deviate only slightly from a Gaussian, in comparison to that of a perfect Gaussian beam. Furthermore, the Z scan obtained by using a particular Gaussian-Bessel beam was found to increase the sensitivity of the Z scan by a factor of 40 and more. One can also envision applications toward optical limiting, for example, by optimizing the roles of self-enhancing internal refractive and absorptive nonlinearities in a similar manner to that shown in Ref. [16]. We anticipate and hope that this theoretical study will prompt an immediate experimental investigation.

S. Hughes thanks the Japanese Society for the Promotion of Science (JSPS) for financial support. J. M. Burzler would like to acknowledge financial support from the German Studienstiftung Cusanuswerk.

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