Enhancement in Reflection Z-scan sensitivity applied to optical crystals

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Recebido em 5 de Maio 1998

We present a new experimental technique for measurements of optical nonlinear effects on surfaces of absorptive and transparent materials. This technique is similar to the RZ-scan technique recently proposed, but uses an incidence angle close to the Brewster angle for a polarized laser beam. A sensitivity enhancement of more than 30 times from the perpendicular incidence case is experimentally observed.

Apresentamos uma nova técnica experimental para medida de efeitos óticos não-lineares em superfícies de materiais absortivos e transparentes. Esta técnica é semelhante à técnica de RZ-Scan recentemente desenvolvida, porém emprega um feixe polarizado com um ângulo de incidência próximo ao ângulo de Brewster. Observamos experimentalmente um aumento de 30 vezes na sensibilidade comparado à técnica com incidência perpendicular do feixe.

The well-known Z-scan technique, introduced by Sheik-Bahae in 1989 [1,2] has been widely used for measurements of optical nonlinearities because of its simplicity and high sensitivity. Several further extensions of this technique have been developed [3,4,5,] and applied to different transparent materials. Recently, the reflection Z-scan (RZ-scan) method has been proposed to investigate highly absorbing materials [6,7] in which the negligible transmitted beam cannot be studied by the transmission Z-scan (TZ-scan) method. The RZ-Scan method, besides its applicability to transparent materials with small absorption, is a useful tool to study surface properties of high-absorbing materials, like semiconductors.

In RZ-scan the nonlinear refractive index is obtained by measuring the wavefront distortion of a laser beam due to the photo-induced modification in the reflection coefficient. This modification is smaller than the corresponding one in conventional TZ-scan, where the laser propagates along all the sample's length, and in many cases is below the measurement sensitivity. This imposes severe limits to the applicability of the RZ-scan technique, since often there are practical limits to the available pump intensities. Moreover, for saturable absorptive media the nonlinear effect is limited due to the saturation of the excited state population.

We present one new method for measuring the nonlinear refractive index and nonlinear absorption in surfaces and interfaces of opaque and transparent samples with an enhanced sensitivity compared to the RZ-Scan technique. This method uses the fact that the p-polarized reflected beam intensity is reduced near the Brewster angle and, as a consequence, the relative variation of this component is highly amplified. The proposed measurement scheme is shown in fig. 1. The Gaussian beam from an Argon laser (514 nm) is focused by lens L3 on the sample surface S at a non-zero incidence angle q. The sample and a folding mirror M move together along the beam direction z, and the reflected beam is measured by detector D. A pair of lenses L1, L2 provides a small waist where a mechanical chopper is located. The laser polarization is parallel to the incidence plane within a precision of 0.5°.

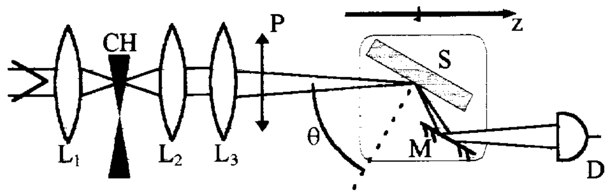

Figure 1. Enhanced RZ-Scan set-up: L1, L2, L3: lenses; CH: mechanical chopper; D: detector; M: mirror; S: sample; P: polarization direction of the beam; q: incidence angle.

The amplitude reflection coefficient of the p-polarized beam for a medium with a complex refractive index $\tilde{n}$ is given by the well-known Fresnel expression [8]:

$$r(\theta) = \frac{\tilde{n}^2\cos\theta - \sqrt{\tilde{n}^2 - \sin^2\theta}}{\tilde{n}^2\cos\theta + \sqrt{\tilde{n}^2 - \sin^2\theta}} \tag{1}$$

Let us consider a medium with Kerr or thermal nonlinearities, in which the modification $\Delta\tilde{n}$ in the nonlinear index depends on the intensity $I : \tilde{n} = \tilde{n}_0 + \Delta\tilde{n}(I)$, where both the linear and nonlinear terms $\tilde{n}_0$ and $\Delta\tilde{n}$ have a real dispersive part n and an imaginary absorptive part k.

In a Gaussian beam the intensity I depends on the distance z from the beam waist position along the propagation direction, and on the radial position ρ across the beam as $I(\rho, z) = I_0[w_0/w(z)]^2 \exp(-2\rho^2/w(z)^2)$, where w_0 is the beam waist and $w(z) = w_0[1 + (z/z_0)^2]^{1/2}$. To evaluate the reflected beam intensity, we expand the reflection coefficient in first order as $r(\theta) = r_0(\theta) + \Delta\tilde{n}\ \partial r(\theta)/\partial\tilde{n}$, where r_0 is the linear term of the reflection coefficient and $(\Delta\tilde{n}\ \partial r(\theta)/\partial\tilde{n})$ is the nonlinear term.

The normalized power reflection R is defined as the ratio between the reflected beam power with and without the nonlinear effect:

$$R(z, \theta) = \frac{\int_0^\infty |r(\theta)|^2 I(\rho, z)\rho d\rho}{\int_0^\infty |r_0(\theta)|^2 I(\rho, z)\rho d\rho} \tag{2}$$

For saturable absorbers the refractive index change is given by [9] $D\tilde{n}(I) = \tilde{n}_2 I/(1 + I/I_s)$, where I_s is the saturation intensity, and the normalized reflection can be expressed by:

$$R(z\,\theta) = 1 + 2\ Re[\hat{r}(\theta)]\frac{4I_s}{I_0^2 w_0^2}\int_0^\infty \frac{I^2(\rho, z)}{I(\rho, z) + I_S}\rho d\rho \tag{3}$$

where the normalized nonlinear reflection coefficient $\hat{r}(\theta)$ is the ratio between the nonlinear and the linear contributions of the reflection coefficient at $z = 0$ and $r_0 = 0$:

$$\hat{r}(\theta) = \frac{\tilde{n}_2 I_0}{r_0(\theta)}\frac{\partial r(\theta)}{\partial\tilde{n}}\bigg|_{\Delta\tilde{n}=0} \tag{4}$$

In non-saturable media, where the refractive index change is proportional to the intensity ($\Delta\tilde{n}(I) = \tilde{n}_2 I$), eq. (3) is reduced to the simpler form $R(z, \theta) = 1 + Re[\hat{r}(\theta)]/[1 + (z/z_0)^2]$.

For simplicity we have approximated the gaussian beam wavefront as a plane wavefront. Considering that the wavefront curvature is smaller than 0.5° for typical values of z_0 and λ, if we measure the nonlinear term of reflection more than 0.5° apart from the Brewster angle the error in such approximation is smaller than 10%. From eq. (4) it is clear that for measurements closer to Brewster angle the nonlinear term in normalized reflection will be increased. But we cannot make these measurements too close to this situation, due to wavefront curvature. We must also consider the problem caused by misalignment of the polarization with the incidence plane. This will introduce an undesirable offset in the reflected intensity. Anyway, in the range we suggest, we can achieve an enhancement factor of 30 times compared to the results for normal incidence, without necessity of more complex description of the nonlinear terms in reflection.

Hence, measuring the reflected power R for different positions z and fitting the results with eq. 3, we can determine the normalized reflection coefficient $\hat{(\theta)}$ and the nonlinear index n_2. At normal incidence ($\theta = 0$)

this corresponds to the technique proposed in [6], in which case the normalized reflection coefficient is given by $\hat{r}(0) = 2\tilde{n}_2 I_0/(\tilde{n}_0^2 - 1)$.

We have applied the proposed scheme for a Gadolinium ($GdAlO_3$:Cr^{+3}) crystal, with linear refraction and extinction coefficients $n_0 = 2.0$ and $k_0 = 6 \quad 10^{-6}$. This crystal is transparent, allowing a direct comparison with the standard TZ-scan technique. In order to eliminate the linear contribution of the reflected beam [10], we chop the incident beam and analyze the time evolution of the reflected signal. Immediately after the chopper is opened the excited state population is very small and the refractive index is purely linear. As this population increases so does the nonlinear index, until a saturation is achieved. Hence, dividing the final value by the initial one we can eliminate the linear contribution of the refractive index and directly obtain $R(z, \theta)$.

We have made the time resolved measurement with the sample fixed at the waist. By tilting the sample close to the Brewster angle $\theta_B = 63.4°$ the value of $|\hat{r}(\theta)|$ is increased, making the nonlinear effect measurable, as shown in Fig. 2. In Fig. 2a the incidence angle was smaller than the Brewster angle, which implies that the reflection coefficient increases with the nonlinear effect. For incidence angles bigger than θ_B, the reflection coefficient decreases with the nonlinear effect, and the time evolution shows a decreasing curve for $R(t)$ as we can see in Fig. 2b.

The asymptotic value in Fig. 2 give us $R(z = 0, \theta)$, from which n_2 can be obtained. We have used the saturation intensity value [9] $I_S = 1600$ W/cm^2 to obtain the nonlinear index value $n_2 = (4.4 \pm 0.5) \times 10^{-8}$ cm^2/W. At normal incidence the time evolution amplitude shown in Fig. 2a is 25 times smaller, which means that it would be very difficult or impossible to measure this variation with the RZ-Scan.

In order to show the dependence of the enhancement with the tilting angle, we have performed several measurements like the one of Fig. 2 for different angles, as shown in Fig. 3. As we approach a range 0.5^0 apart of Brewster angle from both sides in the figure, we see a big enhancement in the nonlinear effect, close to 30 times the one obtained for normal incidence. Fitting $R(0, \theta)$ to the experimental data of Fig. 3 (continuos line), we obtain $n_2 = (4.2 \pm 0.5) \times 10^{-8}$ cm^2/W.

Another way of performing the n_2 measurement is to keep the incidence angle fixed at a value close θ_B and perform a tilted RZ-scan. The result obtained is shown in Fig. 4. In this figure each point is an average of 250 points obtained by the temporal evolution as in Fig. 2. The time evolution os used here only as a normalization method to increase the set up sensitivity. The continuous line in this figure shows the fitted theoretical expression for $R(z, \theta)$, equation (3), from which the value $n_2 = (4.0 \pm 0.4) \times 10^{-8}$ cm^2/W is obtained.

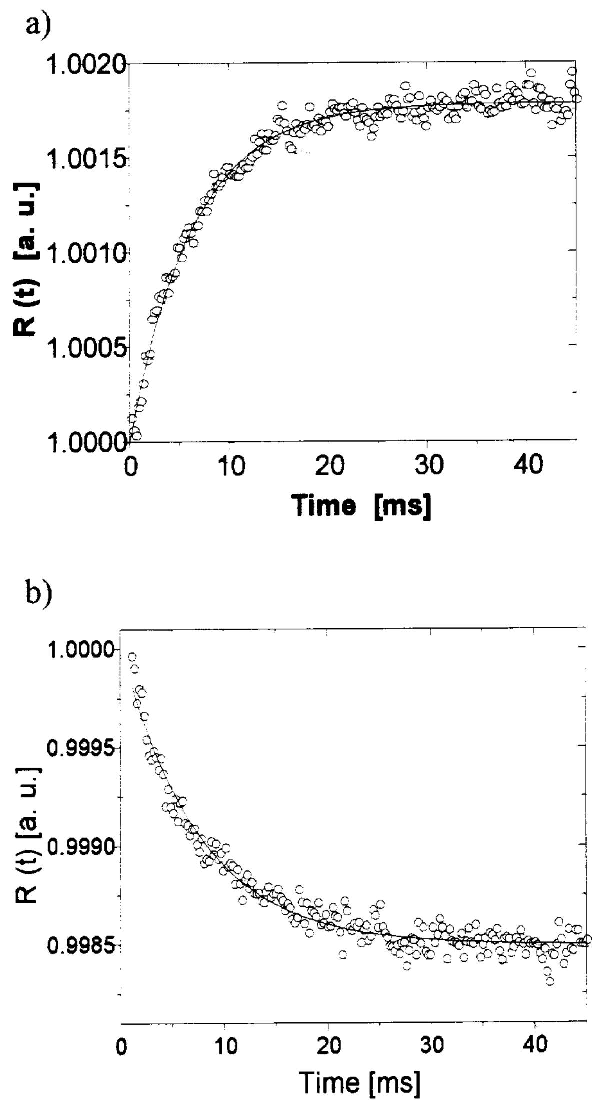

Figura 2. Time evolution of the reflected beam for a) $\theta = 62.6^0$ and b) $\theta = 64.1^0$. Each point is an average over 1000 cycles of the chopper. $I_0 = 10$ kW/cm^2, $w_0 = 50\mu$m.

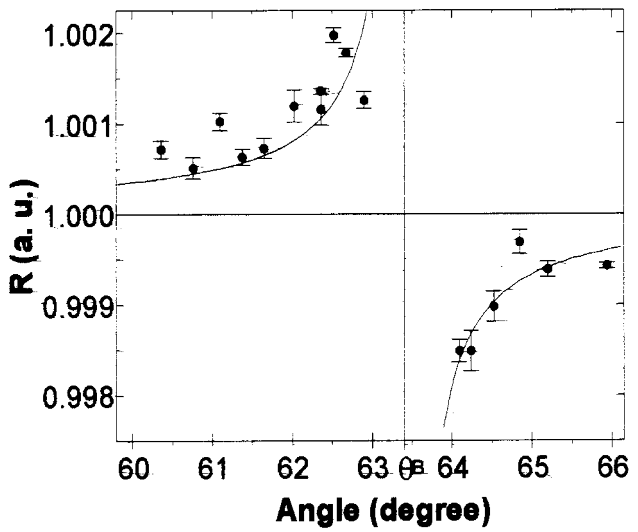

Figura 3. Sensitivity enhancement for different incidence angles. $I_0 = 10$ kW/cm^2, $w_0 = 50$ μm.

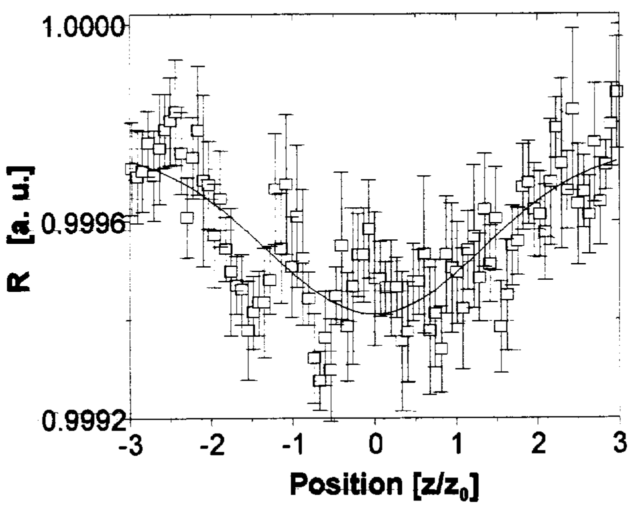

Figura 4. Tilted RZ-Scan measurement, performed with an incidence angle $\theta = 65^0 \cdot I_0 = 10$ kW/cm^2, $w_0 = 50\mu$m.

The values of n_2 obtained from these measurements agree with that measured by the conventional TZ-Scan technique within the experimental errors. Those results show that our method can consistently be used to measure small variations in the refractive index. This method may be applied for both optical crystals and highly absorbing materials like semiconductors or doped glasses. The method presented here enables us to study nonlinear effects in surfaces and interfaces with a simple single beam technique with high sensitivity. We can also use this technique to distinguish surface from bulk nonlinear effects.

In the same way as in the RZ-scan technique, this method can be used also to determine the nonlinear absorption coefficient. In order to do so, we should place an aperture in front of the detector, converting by this way phase variations in the reflected beam into intensity variations as in the usual TZ-Scan method.

The technique demonstrated here gives a 30 times enhancement in the observable nonlinear effect in the reflection. Higher enhancements can be obtained, since we take care about the plane wave aproximation. Considering the curvature of the beam wavefront, we must consider the reflection dependence on the incidence angle in equation (4). If we work in a region where the change of $\hat{r}(\theta)$ is small within the curvature angle, we can use the simple plane wave aproximation sugested here.

The authors acknowledge the financial support from Fundação de Amparo à Pesquisa do Estado de São Paulo (FAPESP) and Fundação Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES).

References

1. M. Sheik-Bahae, A. A. Said and E. W. Van Styland, Opt. Lett., **14**, 955 (1989).
2. M. Sheik-Bahae, A. A. Said, T. Wei, D. J. Hagan, and E. W. Van Styland, IEEE J. Quantum Electron., QE-**26**, 760 (1990).
3. H. Ma, A. S. L. Gomes and Cid B. Araújo, Appl. Phys. Lett., **59**, 2666 (1991).
4. M. Sheik-Bahae, J. Wang, R. DeSalvo, D. J. Hagan and E. W. Van Styland, Opt. Lett., **17**, 258 (1992).
5. W. Zhao and P. Palffy-Muhoray, Appl. Phys. Lett., **63**, 1613 (1993.
6. D. V. Petrov, A. S. L. Gomes and Cid B. Araújo, Appl. Phys. Lett., **65**, 1067 (1994).
7. D. V. Petrov, A. S. L. Gomes and Cid B. Araújo, Opt. Commun., **123**, 637 (1996).
8. M. Born and E. Wolf, *Principles of Optics* (Pergamon Press, London, 1959) p. 614.
9. T. Catunda and L. A. Cury, J. Opt. Soc. Am. B, **7**, 1445 (1990).
10. L. C. Oliveira and S. C. Zilio, Appl. Phys. Lett., **65**, 2121 (1994).