Influence of strong light beams on the nonlinear refraction and absorption coefficients of transparent materials: supplementary material

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ABSTRACT

In this supplementary material we briefly describe how to adapt the expressions in Amaral et al., J. Opt. Soc. Am. B 35, 2977-2985 (2018), to the situation described in the main manuscript.

**Keywords**: third order nonlinear measurement, Z-scan, stimulated light scattering, CS2

# Introduction

To convert the expressions in Ref. [1] to the present manuscript, it is assumed for simplicity that the photon number is proportional to the power and the intensity, such that $n=cI$. We also define the threshold intensity $I\_{th}$ for the onset of the spatial energy redistribution and $I\_{δ}=\left(γ'L\_{eff}\right)^{-1}$ as the typical intensity scale where the energy transfer between the initial wave and the scattered waves becomes efficient.

We use the simplest model, where only the nonlinear (NL) scattering is relevant. We denote $I\_{l}$ as the intensity of the wave along the initial wave direction and $I\_{s}$ as that due to the scattered light. According to the WCM they must satisfy

$$I\_{l}=I\_{i}\frac{{I}/{I\_{th}}}{{I}/{I\_{th}}+e^{{\left(I-I\_{th}\right)}/{I\_{δ}}}},\left(1\right)$$

$$I\_{s}=I\_{i}\frac{e^{{\left(I-I\_{th}\right)}/{I\_{δ}}}}{{I}/{I\_{th}}+e^{{\left(I-I\_{th}\right)}/{I\_{δ}}}}.\left(2\right)$$

Only a fraction $f$ of the scattered light power is detected, such that the intensity of the detected light can be represented as $I\_{d}=I\_{l}+fI\_{s}$. $f$ depends on the physics of the energy spatial redistribution due to the NL response and of the experimental detection geometry. Specifically, for the NL transmittance experiment in Fig. 4, it can be stated that the detected intensity varies according to

$$T\left(I\right)=1-\left(1-f\right)\frac{e^{{\left(I-I\_{th}\right)}/{I\_{δ}}}-e^{-{I\_{th}}/{I\_{δ}}}}{{I}/{I\_{th}}+e^{{\left(I-I\_{th}\right)}/{I\_{δ}}}},\left(3\right)$$

 which is equivalent to the term between square brackets in Eq. (15) of Ref. [1].

Meanwhile, Fig. 3 contains the NL transmittance when the sample position $z$ changes along the propagation axis. The beam used in our experiments was produced by the transmission of light through a circular aperture (object in Fig. 1) whose radius is $r=1,46$ mm. Near the focus the beam has a Bessel-like transverse profile, and the NL response can be mainly associated with the intense central lobe. To model the beam intensity, we considered that the central lobe beam intensity varies according to

$$I\left(z\right)=\frac{I\_{0}}{1+\left({z}/{z\_{0}}\right)^{2}}\left(4\right)$$

To obtain an estimate of the Rayleigh length of the intense central region, we used that the first ring of zero intensity in the focal plane has $w\_{0}=50μ$m radius. A multimode Gaussian beam whose propagation factor is $M^{2}$ has a Rayleigh range $z\_{0}=π{w\_{0}^{2}}/{\left(M^{2}λ\right)}=14.8{mm}/{M^{2}}$. For a Gaussian beam with a single spatial mode, $M^{2}=1$. From a numerical simulation, we have found that the on axis intensity is divided by 2 after propagation over $8.7$~mm, which indicates that $M^{2}≈1.7$. The definition of an effective Rayleigh length to use in a NL interaction is somewhat problematic for the beam we used. For definiteness and simplicity, we approximate that $M^{2}=2$, such we effectively used that $z\_{0}=7.4$ mm as a reference value for a qualitative agreement between the experimental data and the WCM.

The NL extinction coefficient has a behavior similar to Eq. [16] in Ref. [1], which becomes in the present notation

$$C3=α'\left(1-f\right)\frac{e^{{\left(I-I\_{th}\right)}/{I\_{δ}}}-e^{-{I\_{th}}/{I\_{δ}}}}{{I}/{I\_{th}}+e^{{\left(I-I\_{th}\right)}/{I\_{δ}}}},\left(5\right)$$

 where $α'$ indicates the scaling of the loss coefficient. Since the expressions used in Fig. 2(b) depend on the product of $α'$ and $f$, it was necessary to obtain $f$ in Fig. 3(b) and later use Fig. 2(b) to evaluate $α'$.

Finally, the data in Fig. 2(a) can be described using $η\_{2s}$ only (Ref. [1] notation), or in the present paper notation,

$$n\_{2}\left(I\right)=\frac{n\_{2,scale}}{{I}/{I\_{th}}+e^{{\left(I-I\_{th}\right)}/{I\_{δ}}}}\left[{I}/{I\_{th}}+\frac{e^{{\left(I-I\_{th}\right)}/{I\_{δ}}}-e^{-{I\_{th}}/{I\_{δ}}}}{{I}/{I\_{δ}}+e^{\left(I-I\_{th}\right)}{I\_{th}}/{I\_{δ}}}\right]\left(6\right)$$

# References

[1] Anderson M. Amaral, Albert S. Reyna, Edilson L. Falcão-Filho, and Cid B. de Araújo, “Effective model for nonlinear refraction and extinction coefficients in the presence of stimulated light scattering,” J. Opt. Soc. Am. B 35, 2977-2985 (2018).