Observation of conical emission from dimethylsulfoxide

Viviane Pilla, Leonardo de S. Menezes, Márcio A. R. C. Alencar, Cid B. de Araújo
Departamento de Física, Universidade Federal de Pernambuco
vpilla@df.ufpe.br

Abstract

In this work we studied the effect of conical emission due to Cross Phase Modulation (CPM) in dimethylsulfoxide (DMSO). The pump beam used was obtained from a Q-switched Nd:YAG laser (1064 nm, 9 ns, 5 Hz) and the probe beam was provided by a dye laser operating at 597 nm, excited by the second harmonic of the Nd:YAG laser. Both beams were linearly polarized and carefully overlapped and focused in the sample with a 10 cm focal length lens. When both beams were incident on the sample, it was observed a ring formation around the dye laser beam in the far field region. The conical emission is attributed to the nondegenerate third order susceptibility, \( \chi^{(3)} \) (-\( \omega_1 \); \( \omega_2 \), -\( \omega_2 \), \( \omega_1 \)) and thermal effects. Its measurement was made using the two-color Z-scan technique and the values obtained for \( n_2' \) in the perpendicular and in the parallel orientation of the excitation and probe beam fields are \( \approx 10^{-15} \) cm\(^2\)/W. With basis on these results we calculated the ring pattern formation using the Fresnel-Kirchhoff diffraction integral.

Introduction

A Kerr medium in presence of a laser beam with spatial intensity profile I(r) presents a refractive index profile expressed as \( n(r)=n_0+n_2I(r) \), where \( n_0 \) is the linear refractive index and \( n_2 \) is the nonlinear complex refraction coefficient. Thus, effects due to the Kerr nonlinearity such as self-focusing (\( n_2 > 0 \)), self-defocusing (\( n_2 < 0 \)), self-trapping and cross-phase modulation (CPM) are responsible by a spatial and / or spectral modification of the laser beam due to \( n_2 \). In the case of CPM, the phase of a weak probe beam is affected by a copropagating or counterpropagating pump beam and this effect normally induces the spatial redistribution of the probe beam energy [1, 2]. Theses effects are dependent on the beam’s polarizations, wavelength and spatial-temporal distributions of the energy.

The total phase variation, using the pulsed laser beam, due to effects of the CPM is expressed in terms of time-averaged phase variation in the form:

\[
<\Delta \phi> = \Delta \phi_K + \Delta \phi_{TH}
\]

where \( \Delta \phi_K \) and \( \Delta \phi_{TH} \) are the Kerr and thermal phase variations, respectively, given by [3, 4):

\[
\Delta \phi_K = \frac{n_2}{\sqrt{1.5}} k \int_0^L |E_{\text{pump}}(\rho_1, z)|^2 \, dz
\]

and

\[
\Delta \phi_{TH} = \frac{\alpha \tau_p}{2\rho_0 C} \int_0^L |E_{\text{pump}}(\rho_1, z)|^2 \, dz
\]

where \( \rho_1=r/\omega_1 \), \( x=z/z_0 \) (being \( z_0 \) the confocal parameter), \( k=2\pi/\lambda_p \) (\( \lambda_p \) is the probe beam wavelength), \( \alpha \) is the absorption coefficient, \( \tau_p \) is time of duration of the laser beam, \( \rho_0 \) is the density (g/cm\(^3\)) of the Kerr medium, \( C \) the specific heat (J/gK) and dn/dT the thermo-optic coefficient. The electric field amplitude is expressed as:

\[
e_p(\rho_1, z)=e_{p0}(\rho_1) \exp(-\frac{\alpha}{2}z)
\]

In this way, the probe beam in the sample, can be expressed [3]:

\[
e_p(\rho_1) \propto \exp(-\rho_1^2(1+ix) -i\Delta \phi_K -i\Delta \phi_{TH})
\]

It is important to remember that the understanding of these phase modulation effects can be used to manipulate the optical beam the pulse duration in the time and/ or spatial domain to correct distortion of the beam. Recently, these effects have been used for spectroscopic applications and in the search for photonic devices.
The two color Z-scan technique (TCZST) was employed to study the CPM effect [5, 6]. The empirical expression of the TCZST relate the difference between the normalized peak and valley transmittances (Figure 2(a)) with parameter \( n_2' \) and \( \frac{dn}{dT} \), in the form:

\[
\Delta T_{pv} \approx 0.42 (1-S)^{0.35} \langle \Delta \phi \rangle
\]

where \( \langle \Delta \phi \rangle = k \gamma l_{ex} L_{eff} \), with \( \gamma \) given by:

\[
\gamma = \frac{n_2'}{\sqrt{1.5}} + \left( \frac{\alpha_0}{2pC} \right) \frac{dn}{dT}
\]

In this case, \( l_{ex} \) is the intensity (W/cm\(^2\)) of the excitation beam, \( L_{eff} = (1-e^{-\alpha L})/\alpha \) and \( L \) the sample thickness (cm). Figure 2(a) allows to determine \( z_0 \), since \( \Delta Z_{pv} = 1.7 z_0 \). The Z-scan curve, for materials that present only electronic effects with \( n_2' < 0 \), shows a maximum transmittance (peak (1)), followed by a minimum transmittance (valley (2)). The inverse is expected for samples presenting \( n_2' > 0 \). The real part of the susceptibility \( \chi^{(3)} \) can be found using the expression [7]:

\[\text{Re}[\chi^{(3)}] = \frac{4}{3} \varepsilon_0 \cdot c \cdot n_0^* \cdot n_2' \]

where \( \varepsilon_0 \) is the vacuum electric permittivity and \( c \) the light velocity in vacuum.

The study of the CPM phenomena was made numerically using the Fresnel-Kirchhoff diffraction integral (IDFK) [8]. Initially, let us consider the diffraction of a monochromatic plane wave linearly polarized, in a finite hole in \( S \) plane (described by the coordinates \((x_i, y_i, z)\)) spreading in the direction given by the unit vector. The complex amplitude of the field \( \varepsilon(r_2) \) at point \( P_2 \) in the observation plane \( P \) (coordinates \((x_2, y_2, z + d)\) located at a distance \( d_0 \) of the away from the sample) is given by the sum of the field \( \varepsilon \) due to all the points \( P_1 \) of the plane \( S \). In cylindrical coordinates \((r_i^2 = x_i^2 + y_i^2, i = 1 \text{ or } 2)\) the IDFK, can be written in the form [8]:

\[
\varepsilon(r_2) = \frac{ik}{d} \text{Exp} \left( -ik \left( d + \frac{r_2^2}{2d} \right) \right) \int_0^\infty \text{Exp} \left( - \frac{ikr_1^2}{2d} \right) \varepsilon_s(r_1) J_0 \left( \frac{kr_1r_2}{d} \right) r_1 \text{dr}_1
\]

**Results**

A glass cuvette 2 mm thick containing DMSO was placed in the focal region of an 10 cm focal length lens. The formation of one ring in the transversal profile of the probe beam was observed in the far field. Both copropagating beams were linearly polarized, carefully overlapped and focused on the sample.

The experimental results of CPM effect for the DMSO solvent are shown in Figure 1. The excitation and probe beams used were perpendicularly polarized. A LBA-PC Spiricon CCD detector was placed at approximately 15 cm after the sample. The CCD dimensions were 3.6 mm x 4.9 mm. The measurements were performed using optical filters to block the IR beam, allowing only the probe beam to pass. Neutral density filters were used to prevent the saturation of the detector in the central spot region of the probe beam applied at the recorded image in order to improve the ring image around the central profile of the laser beam.

**Figure 1:** CPM effect in DMSO using a dye laser (597 nm, intensity lower than \( 4 \times 10^6 \) W/cm\(^2\)) as a probe beam: (a) excitation beam absent and (b) when the pump beam (1064 nm, \( l_{ex} = 4 \times 10^8 \) W/cm\(^2\)) is present with polarization perpendicular to the probe beam. The ring structure is clearly seen around the Gaussian profile of the probe beam.
Figure 2(a) presents typical TCZST measurements in DMSO. It was obtained using the same laser setup described above. From these curves we obtain $\Delta T\text{pv}$, which is related to $\gamma$. In Table 1 are presented the results of $\gamma$ for the DMSO for the parallel ($\|\$) and perpendicular ($\perp\$) polarization of the excitation and probe beams. This parameter is related to $n_2$ and to the thermal parameter $dn/dT$. Taking into account that the thermal signal reaches its maximum more than 40 ns after the heat delivery we estimated that no more than 20 % of maximum thermal value could be in effect during the pulsewidth [9]. Using Eq. (7) and the thermal parameter obtained from the literature for DMSO [10], we can evaluate the value of $n_2$. We used in our calculations the values: $\rho = 1.1 g/cm^3, C = 1.96 J/gK$ e $dn/dT = 3.58 \times 10^{-4} K^{-1}$.

**Figure 2:** (a) TCZST measurements in DMSO ($L = 2 \text{mm}$). $I_{exc} = 2 \times 10^9 \text{W/cm}^2$ (1064 nm), parallelly polarized to the probe beam (597 nm), which had an intensity lower than $4 \times 10^6 \text{W/cm}^2$ (b) Temporal response of the probe beam passing through the sample, while the pump beam is also present ($\perp$ polarization) at positions (1), (2) e (3). It was not observed any variations in the temporal response that was kept about 11 ns.

Relative polarization between pump and probe beams | $\gamma$ (10^{-14} cm^2/W) | $n_2$ (10^{-15} cm^2/W) |
--- | --- | --- |
$\|$ | $-1.2 \pm 0.2$ | 5.9 |
$\perp$ | $-2.1 \pm 0.9$ | -3.8 |

**Table 1:** Two color Z-scan results in DMSO, using $\lambda_{exc}=1064$ nm and $\lambda_{probe}=597$ nm. $\gamma$ is the experimental parameter obtained from TCZST. $\tau_e$=9 ns and $\alpha=(0.12 \pm 0.04) \text{ cm}^{-1}$

After the two color Z-scan measurement had been performed, the positions of the peak (1), valley (2) and far from focus (3) were identified (Figure 2(a)). The sample was placed in each of these positions and time evolution measurements of the probe beam’s intensity central profile were made (Figure 2 (b)). It was observed that the temporal responses were coincident, with in $\pm 2 \%$ accuracy. These results indicate that the origin of the CPM effects is not thermal, as pointed out on reference [11]. Measurements of the dependence of the ring intensity with the pump and probe beams intensity were also made. In order to perform this experiment a photomultiplier tube coupled to an optical fiber of 100 $\mu$m in diameter was placed in the focal position of a convergent lens, located in far field (after the cuvette containing DMSO). A circular mask was put at the center of this lens in order to block the probe beam spot and to allow only the ring signal to be collected. It was observed that the ring intensity was quadratically dependent on the excitation beam intensity and varies linearly with the probe beam intensity. These results were confirmed for both polarization cases, and indicate that the ring formation observed is a third order process.

Using the Eqs. (1-5, 9) is possible to evaluate the intensity profile of the probe beam $I(p)\equiv|\rho(p)|^2$ on the far field. Figure 3 presents the probe beam intensity profiles, taking into account the Kerr and the thermal effects. All the curves are normalized to the value of the maximum point for $<\Delta \phi>= 0$. It is possible to observe that for $I > 8 \times 10^6 \text{ W/cm}^2$ the ring formation occurs around the probe beam central profile. This result is in agreement with the experimental results, where for $I > 5 \times 10^6 \text{ W/cm}^2$ is possible to measure this ring formation.
Figure 3: Numeric calculation of $I(\rho_2)$ as a function of $\rho_2 = \tau_2/w_2$, using the phase variation $\Delta \phi_K + \Delta \phi_{TH}$, $\gamma = 2.1 \times 10^{-14}$ cm$^2$/W, $\alpha = 0.12$ cm$^{-1}$, $dn/dT = -3.58 \times 10^{-4}$ K$^{-1}$ [10], $\rho = 1.1$ g/cm$^3$, $C = 1.97$ J/gK [10], $w = 0.0025$ cm and $\tau_p = 9$ ns.

Conclusions
Cross Phase Modulation effects (CPM) have been studied in DMSO. Experimental results using two color and transient Z-scan techniques show that the conical emission can be attributed to the third order nondegenerate susceptibility $\chi^{(3)} (-\omega_1; \omega_2, -\omega_2, \omega_1)$ and thermal effects. The TCSZT was employed to determine $n_2$ (the thermal parameters of DMSO are known). Our results were $n_2 \approx 10^{-15}$ cm$^2$/W to the parallel and perpendicular relative polarizations between the probe and pump beams. Based on these results, we calculated the ring patterns using the Fresnel-Kirchhoff diffraction integral and conclude that the contribution of the two effects, electronic and thermal, are responsible for ring formation in the profile of the probe laser beam.

Acknowledgements
We acknowledge the support of the Brazilian agency Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq).

References