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Study of multiphoton absorption processes in a perylenediimide derivative using thermal lensing technique

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Two multiphoton absorption processes were revealed in a perylenediimide derivative, at the wavelength of 1064 nm, by using the thermal lens technique. One of the observed multiphoton processes is two photon absorption (TPA) while the second is a four photons process. The value of the measured TPA coefficient is $\beta = 0.12\ 10^{-10}$ cmW^{-1}

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1. Introduction

Organic materials that exhibit strong multiphoton absorption properties are currently of great interest because of their potential applications in optical limiting devices, frequency up-converted lasing, three-dimensional optical data storage, multiphoton fluorescence microscopy, photonic crystal fabrication and photodynamic therapy [1].

In this work, we present multiphoton absorption measurements at wavelength of 1064 nm in perylenediimide-Br_2 ($PDIBr_2$) (see structure Fig. 3a) dissolved in chloroform. This compound was recently synthesized for photovoltaic applications [2].

We also present the results obtained in nitrobenzene at the same wavelength, with the aim of comparing different materials.

To study the multiphoton processes induced in the studied materials, we use the pump-probe thermal lens technique which is known for its high sensibility [1,4,5].

Besides, this technique allows to easily distinguish between the various non linear absorption processes present in the material [1] that is a major advantage with regard to the other methods of measurement such as the z-scan technique [7].

2. Experimental details

Nonlinear absorptivities of $PDIBr_2$ and nitrobenzene were measured using the experimental setup depicted below.

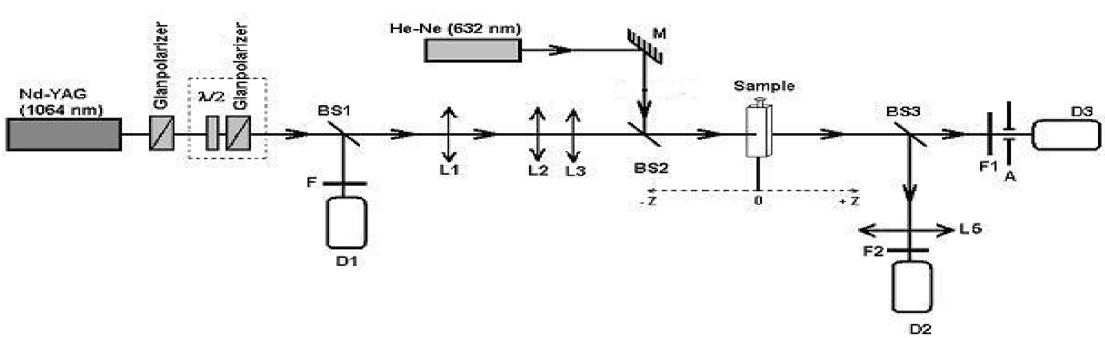


Fig. 1. Experimental set-up: Nd-YAG : pump laser; He-Ne : probe laser, BS : beam splitter, L : lens ; F : filters; A : aperture, D: photodiodes.

The excitation of the samples is provided by 30 ps light pulses at 1064 nm and the probe beam is from a cw He-Ne laser (1mW, 632 nm). The thermal lens signal TLS is defined here as the relative change of the probe beam transmission through a small aperture located in the far field.

The details of the theoretical expression of the thermal lens signal (TLS) S_{TL} can be found in the references [1,4]. For Gaussians beams, S_{TL} is related to the phase shift induced by the thermal effect Φ by the relation :

$$S_{TL} = K.\Phi \quad (1)$$

where K is a coefficient that depends upon geometrical factors like the positions of the beam waists and Rayleigh parameters of the pump and probe light beams.

The phase shift is related to the pump beam energy E_p [1, 4] and we can write:

$$\Phi \propto E_p^n \quad (2)$$

where E_p is the pump beam energy and n is the number of photons absorbed in transition.

The TPA coefficient β, which is a characteristic of the material is given by :

$$\beta = \frac{4.\lambda_{pb}.D.\Phi}{\pi^{3/2}.\tau.\ell.I_p^2.\frac{\partial n}{\partial T}} \quad (3)$$

where D is the thermal diffusivity, τ is the pulse duration, ℓ is the sample path length, Ip is pump beam intensity, λ_{pb} is the wavelength of the probe beam and $\partial n/\partial T$ is the thermal gradient of the refractive index.

3. Results and discussion

The results obtained for the pure solvent and solutions are presented below. Fig. 2 shows the UV- Vis spectra of the chloroform, which is the solvent used in our measurements. Fig. 3 (a) shows the UV-Vis spectrum of the organic material perylenediimide-Br_2 (PDI-Br_2) with the chemical structure and Fig. 3 (b) represents the TLS of the solution (i.e. chloroform + PDI-Br_2) as a function of the incident laser energy.

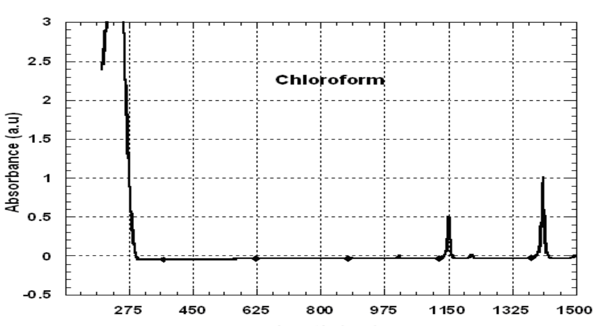
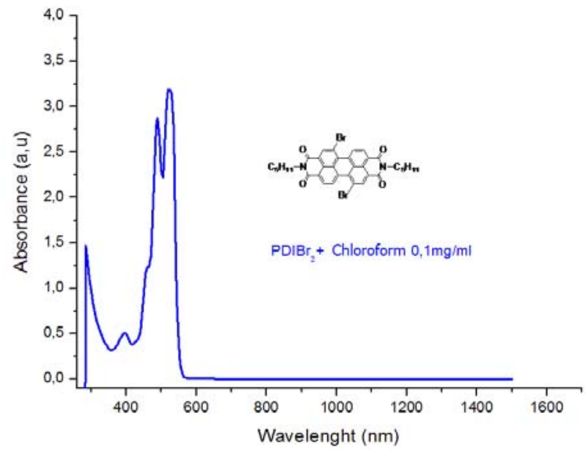
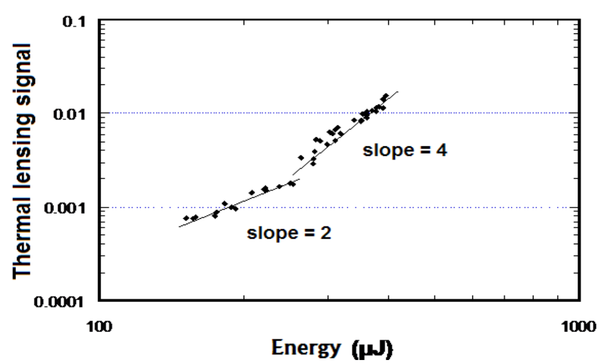


Fig. 2. UV-Vis spectra of chloroform.

Two photons absorption is not expected at this wavelength and that is confirmed by the UV-Vis spectrum that shows no linear absorption in the range 310 nm-1104 nm.



(a)



(b)

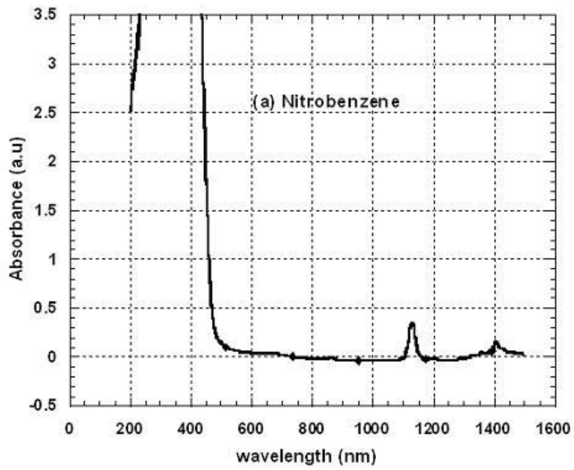
Fig. 3. (a) UV-Vis spectra of PDI-Br_2; (b): TLS as a function of pump beam energy (pump wavelength 1064 nm).

Thermal lens curve reveals the existence of two photon absorption describes by the line of slope 2 and other nonlinear losses or presence of other nonlinear effects described by the second line of slope 4. A slope of 4 was also observed in our measurement at 1064 nm in nitrobenzene.

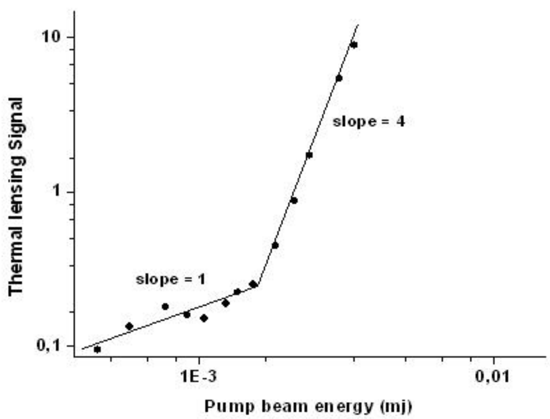
The value of the TPA coefficient for the solution is calculated for the region of slope 2:

$$\beta = 0.12 \times 10^{-10} \text{ cmW}^{-1}$$

Fig. 4 (a) represents the UV-Vis spectra of nitrobenzene and Fig. 4 (b) shows the TLS of nitrobenzene as a function of the pump energy.



(a)



(b)

Fig. 4. (a) UV-Vis spectra of nitrobenzene; (b)TLS of Nitrobenzene as a function of pump beam energy (pump wavelength = 1064 nm).

The four photon phenomenon observed with the wavelength of 1064 nm in solution, can be explained within the framework of infrared multiphotonic photochemistry. Indeed, when the energy of the exciting photon is located in the area of the near infrared, this energy is sufficient to increase the energy of rotation or vibration of a molecule. The molecule "jumps" then from the level of rotation to another or a higher level of vibration of a rotation level [1].

4. Conclusions

This work shows that thermal lens method allows to identify easily many types of nonlinear absorption processes (two photons or multiphoton absorption) and to determine the nonlinear absorption coefficient with greater sensitivity.

On the other hand, the experiments carried out in PDI-Br_2 at the wavelength of 1064 nm reveal the existence of multiphotonic processes in this organic material. Studies of other compounds of the same family are in progress.

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