Article reference: 36123v1

Dear Editor Dr. Shirota,

Thank you very much for the opportunity to review the article entitled "**Effect of femtosecond laser pulse repetition rate on nonlinear optical properties of organic liquids**" by Sandeep K Maurya, Dheerendra Yadav , Debabrata Goswami.

The manuscript describes the effect of the repetition rate of femtosecond laser pulses on the two-photon absorption and nonlinear refraction of pure organic liquids using Z-scan technique. Such a study provides a way to determine the nature of light-matter interaction, explicitly enabling the identification of the linear versus nonlinear regimes. The manuscript identified the thermal load dissipation time for the organic liquids. the experimental results are in good agreement with the theoretically calculated decay time for the dissipation of thermal load.

Overall the manuscript appears to be clearly and carefully written. The results presented are interesting for the field of pump probe technique. I think that the manuscript might deserve publication in Peer J Physical Chemistry after some points are dealt with and some missing details are added prior to publication as follows:

1- *In page # 6, Lines 68-76, Introduction "in turn, depend on the repetition rate of the laser pulses. In the recent past, a vast study on the effect of high repetition rate (HRR) laser pulses have been reported, where thermal load due to a train of laser pulses overwhelm pure single pulse NLO effects. There have been several attempts to obtain the pure NLO property free from thermal inhibition from intense laser pulses. In recent years, optical chopper and pulse picker are being used to manage the cumulative thermal effect in the optical medium [26-29]. However, these techniques have their limitations due to their operating principles. For an instant, use of optical chopper in the determination of NLO property minimizes cumulative thermal effect from intense HRR laser pulses [30] with an enhancement in the signal to noise ratio (SNR) for the nonlinear optical effect."*

In addition to Ref. 26-30, there are more recent work are suggested to be included as "Method for an accurate measurement of nonlinear refractive index in the case of high-repetition-rate femtosecond laser pulses" Journal of the Optical Society of America B 36(5):1264-1251 and "Experimental investigations of nonlinear optical properties of soda-lime glasses and theoretical study of self-compression of fs laser pulses" Optics & Laser Technology 116(0030-3992):276-283.

2- *In page # 7 Lines 107-110, Materials & Methods "Our femtosecond experimental setup involves a tunable repetition rate Ti:Sapphire Regenerative Amplifier System (Spitfire-Pro, Spectra-Physics Inc. USA) that is seeded with a Spectra-Physics Mai-Tai Ti:Sapphire oscillator, having 82 MHz repetition rate with wavelength tunability of 780–820 nm. Figure 1 shows a schematic of the experimental setup, which describes closed".*

Please add more details about the tunable repetition rate Ti: Sapphire Regenerative Amplifier System (Spitfire-Pro, Spectra-Physics Inc. USA) and how to tune the repetition rate from 10 Hz to 1000 Hz ? Also which type of Mai-Tai has been used? The available Mai-Tai cannot provide 82 MHz repetition rate and wavelength tunability of 780–820 nm.

3- *In page # 8 Results and Discussion, Line 152 "76 MHz repetition rate of the laser pulse as shown in Fig. 3. Hence, reversal of sign in the".*

What is the repetition rate? Is it 82 MHz or 76 MHz?

4- *In page # 9 Results and Discussion, Line 189 "with varying repetition rate show a significant change in the peak-valley transmittance in both".*

How significant the change? It is not really clear because all curves displayed in Figure 4 are almost typical.

5- Nothing mentioned in the text about the used mirror in the experimental setup (Figure 1). For example, does the mirror used in the setup have zero dispersion at 800 nm? It is necessary to explain the possible dispersion and how to do the compensation.
6- Please mention the pulse duration in each figure captions
7- Nothing mentioned in the text about the uncertainty in the experimental results, so please discuss this issue in the text.
8- Please discuss in the text the error bar in Figure 8
9- In Figure caption 8 please remove the upper two lines.
10- In the similar experiment that apply the Z-scan, it known that the interplay between self-phase modulation SPM and group velocity dispersion GVD play a

significant role in altering the peak irradiance inside the sample resulting in strongly influenced of the output transmission. What is the contribution of the nonlinearity SPM and GVD in the present setup?

Also here are few typographical errors:

- Line 299, page 13- “Sakakibara, H. Kuroda, Opt. *Comm*. 2004,**231**, 433-438.” should be replace by “Sakakibara, H. Kuroda, Opt. *Comm*. 2004,**231**, 431-436. "
- Page 16 the x-axis of Figure 2 a and b “Sample Position” should be replace by “Sample Position (mm)”

Best regards

Tarek Mohamed

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Tarek Mohamed, PhD
Professor of Laser Physics &
Dean of the Laser Institute for Research and Applications (LIRA),
Beni-Suef University, Egypt
Phone: +201159492205
Email: tarek_mohamed1969@lira.bsu.edu.eg ; tarekhassan1969@gmail.com
Web: http://www.bsu.edu.eg/LIRAHome.aspx?cat_id=46