2024, 21(6): 2137-2145
https://doi.org/10.21123/bsj.2023.8383
P-ISSN: 2078-8665 - E-ISSN: 2411-7986

Baghdad Science Journal

Design and develop an optical setup related to the pump-probe technique for spectroscopic study

Saad Kh. Rahi*[1], ***Muayyed J. Zoory***[1], ***Alaa H. Ali***[2]

[1]Department of Physics, College of Science, Mustansiriyah University, Iraq, Baghdad
[2]Ministry of Science and Technology, Iraq, Baghdad
*Corresponding Author.

Received 17/01/2023, Revised 19/05/2023, Accepted 21/05/2023, Published Online First 20/11/2023, Published 1/6/2024

Abstract

In this research, an optical system using Pump-Probe technology was designed to study the effect of changing the Frequency values, integration time, delay time, and laser pulse width on the pumping pulse energy on an arbitrary laser dye material with an absorption coefficient of about 650 nm. we found that decreasing the pumping time to 10% of the pulse time is not suitable for the pump-probe technique because there is no resonance between the pumping time and the detector response to the source used. Also, we indicate that increasing the pumping time to 20% of the pulse time results in the highest possible detector response at an integration time 150 µs in the pump-probe technique. therefore, we noted that the detector response becomes more suitable for working in the pump-probe technique as the pumping time increases, as well as the efficiency of the detector, which increases significantly as the pumping time increases, as well as the integration time for the same delay time, it is more stable at 20% pumping time and collection time 150 µs at a delay time of 12.4 ns.

Keywords: Delay time, Integration time, Optical detector, Pulse Width, Pump-Probe technology.

Introduction

Atomic photon absorption, electron emission, electron reconfiguration, and similar activities all take place in extremely short amounts of time. All of these activities can happen in only a few picoseconds. Understanding the dynamics of excitement inside the material requires studying and observing these very quick events. The uncertainty in these high-speed process measurements, which may be obtained using a number of different methods, must be less than the femtosecond's time scale. Two of the most well-known of these methods are the Stop- flow technique and the Laser flash. Fast optical diodes and stroboscopes, which only offer time precision on the order of (10^{10}) seconds, have stabilized the accuracy of time for rapid, dynamic research[1]. New methods for measuring time have been created that are significantly different from their forerunners in order to circumvent the issue of detector limits. In addition, it is employed to evaluate the effectiveness of the optical intensity conversion process in nonlinear materials. The laser pump-probe technique is a recent development that involves capturing the laser pulse both before and after it has passed through the non-linear material. Scientific advancements have been made in a wide variety of domains due to the use of pump-probe laser

2024, 21(6): 2137-2145
https://doi.org/10.21123/bsj.2023.8383
P-ISSN: 2078-8665 - E-ISSN: 2411-7986

technology, which is today considered a fundamental tool in the study of all spectrum[2].

The technology of the pump-probe is the simplest experimental method for investigating ultrafast electronic dynamics. It was developed at the end of the 20th century. In this technology, a laser pulse with an ultrashort duration is split into two beams: the first one, which is referred to as the pump, is used to make excitation in the sample, causing a system that is out of balance as a result, and the second one, which is referred to as the probe[3], its principal responsibility is to track the fluctuations in the sample's optical properties that are caused by the pump, such as its reflectivity or transmission, and refractive index, etc. Fig. 1. The pump beam is used to generate changes in the optical behavior, and measuring these changes as a function of the delay time between the arrival of the pump pulse and the probe pulse can provide information about the relaxation of electronic states in the sample[4].

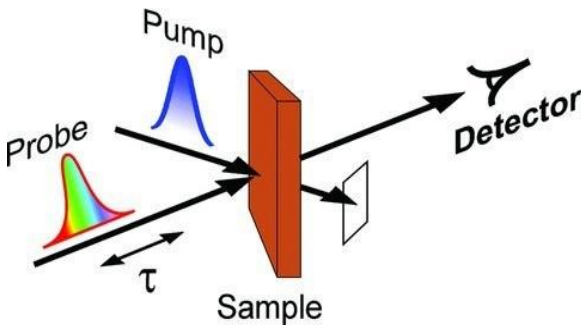


Figure 1. Scheme of a pump–probe experiment[5]

The femtosecond time resolution is ideally suited for the detection of the duration of electronic excitations using the pump probe spectrometer that was discussed above. We can observe the distribution of these excitations in the actual space that they occupy using a technique known as Transient Grating Spectroscopy. This technique requires superimposing two pulses as a femtosecond on the sample in order to generate sinusoidal intensity modulation, which, in turn, causes a density grating of photoexcitation's to occur in the material[4]. The index of refraction of a periodic modulation is created as a consequence of the fact that the index of refraction is dependent on the excitation density of the local area. This is because the refractive index is dependent on the density excitation of the local region. Changing either the laser's wavelength or the angle that splits the two beams will cause a change in the period at which this pattern is repeated in real space. This shift may be caused by either the laser's wavelength or the angle that divides the two beams. As a result, a probe pulse that is incident on this pattern will undergo both reflection and diffraction when it interacts with it. If we measure the temporal, we can calculate both the "reflected" and "diffracted" beams[3].

The phenomenon known as quantum nonlinearity can be traced back to the observation that, when subjected to the stipulation of strong coupling, a system made up of a single atom and a single cavity mode exhibits characteristics that are notably distinct from those of the atom alone (in the absence of the cavity), the cavity alone (in the absence of the atom), or the combination of the two alone. This is due to the fact that quantum nonlinearity results from the fact that, under the condition of strong coupling, a system that is constituted of a single atom and a single electron has the potential to exhibit quantum nonlinear behavior[5]. In fact, the composite system results in the birth of a brand-new quantum entity that is referred to as the atom–cavity molecule. This creature is made up of both matter and light, and it has a spectrum of energy that is completely distinct from any other. This spectrum may be seen as an infinite staircase of paired states that are referred to as "dressed states." Investigating the initial doublet, which has one quantum of energy, may be done using a variety of techniques, one of which is laser spectroscopy. The resulting spectrum, known as the normal-mode spectrum, is independent of the laser's power level during weak probing[6-8]. It consists of a pair of resonances that are symmetrically split between the atomic resonance and the cavity resonance. This spectrum used to be seen with beams of atoms, but recently it has been studied with single atoms held in dipole fields. It is at the center of most cavity quantum electrodynamics (QED) research, even outside of atomic physics, and is used as a benchmark for strong atom-cavity interaction. Keep in mind that a linear dispersion theory or a linked oscillator model may provide a sufficient classical explanation for the normal-mode spectrum by itself (atomic dipole and cavity field). These two models and explanations are equivalent[9].

2024, 21(6): 2137-2145
https://doi.org/10.21123/bsj.2023.8383
P-ISSN: 2078-8665 - E-ISSN: 2411-7986
Baghdad Science Journal

Materials and Methods

Pulse Width Modulation (PWM)

It's a method for making an analog signal out of a digital one. Two major properties of a pulse-width-modulated (PWM) signal are its pulse width and its frequency. A signal's pulse width is the proportion of time it is in its high (on) state relative to the full signal cycle[10]. The frequency of the pulse width modulator is what controls how quickly its high and low states alternate (e.g., 1000 Hz would be 1000 cycles per second). If the digital signal is turned on and off quickly enough and with a high enough pulse width, it can simulate a constant voltage analog signal and be used to supply power to devices. The input signal alternates between on and off at predetermined intervals, altering the power supply of a device. The voltage at the load is directly related to the voltage at the source. The on/off cycle shifts the average signal intensity[11]. Pulse width D, expressed as a percentage of the PWM period, defines the on time (or frequency) relative to the off time (or period) of a PWM signal. The pulse width is described in Eq. 1[11] and shown in Fig. 2.

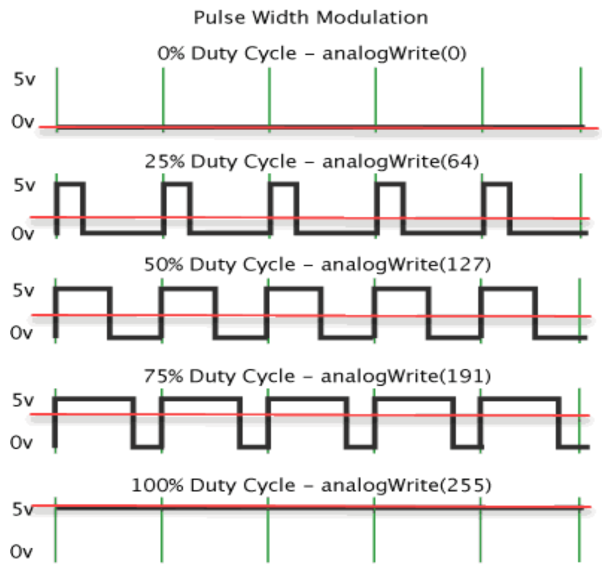


Figure 2. PWM signal [8]

$$\tau = \frac{\tau_i}{f} \times 100\% \qquad 1$$

Where τ is pulse duration(sec), τ_i is percentage ratio and f is frequency.

Some studies have been achieved experimental study for the investigation of pump- probe tech in the nonlinear optical system have been achieved to study the conversion efficiency and generation of the second harmonic at 532 nm in a (KTB) crystal at the fundamental wavelength of a passively (Q-switch) (ND-YAG) laser at an at a wavelength of 1064 nanometers[9].

Two-color pump-probe tech. had been done to study an electronically delayed two-color pump-probe instrument that was developed using two synchronized laser systems. The instrument has picosecond time resolution and can perform scans over hundreds of nanoseconds without the beam divergence and walk-off effects that occur using standard spatial delay systems[12]

The experimental part

The Pump Probe system was designed by using a laser source (**laser diode**) with a wavelength of **650 nm**, a beam splitter, a number of front-coated mirrors, and a spectrometer. The devices were arranged on an optical bench, as shown in Fig. 3.

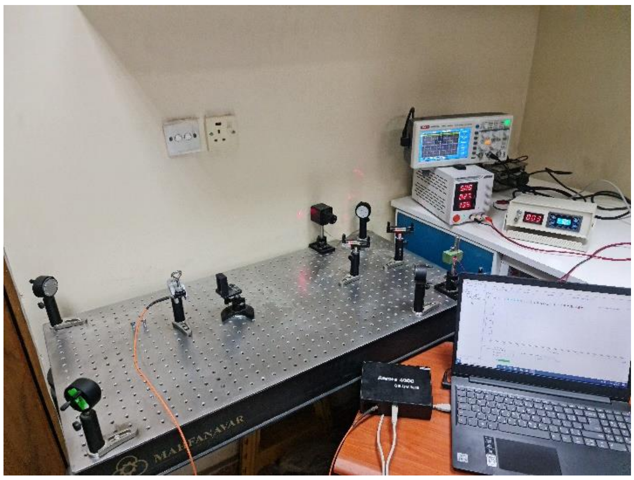


Figure 3. pump probe system

The diagram depicted in Fig. 4 facilitates the identification of the different parts and processes of the experiment. Particularly, the laser pulse is aimed at beam splitter 1, which subsequently divides the resultant laser beam into two equal beams, each comprising 50% of the original beam. One of the beams, commonly referred to as the "pump" beam, is directed towards beam splitter 2. This particular splitter divides the beam into two equal parts, each comprising 50% of the original beam. The main purpose of this beam is to stimulate the atoms that

2024, 21(6): 2137-2145
https://doi.org/10.21123/bsj.2023.8383
P-ISSN: 2078-8665 - E-ISSN: 2411-7986
Baghdad Science Journal

compose the sample material. The resultant beam is pointed to the sample material. The second beam, which is called the "probe" and originates from splitter 1, is tasked with detecting any changes that may take place within the material. This beam travels along an extensive optical path that involves the use of multiple mirrors **M1**, **M2**, **M3**, and **M4** a delay time is generated "t" which varies correspondingly with the distance between the mirrors. Subsequently, the beam is directed towards beam splitter 2 and subsequently towards the sample material.

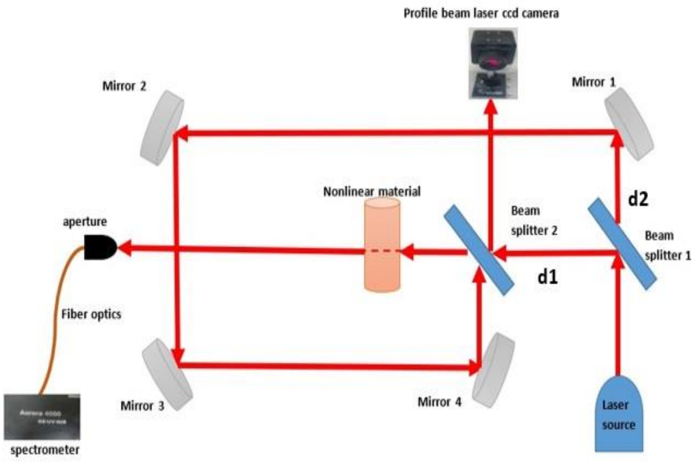


Figure 4. A scheme for the work of the system that was designed for the (pump-probe) technique

The used laser was a continuous laser (**c.w laser**) and it was converted into a pulsed mode using the **(PWM)** Fig. 4. pulse width modulator circuit, it can be controlled to modulate the frequency of the laser pulse from 1 Hz-150 KHz, and also control the pulse width of the laser pulse from 1%-99% of the original pulse width. By using an oscilloscope of type ((UNI-T) 200 MHz with two channels storage mode), the electrical pulse generated by PWM was verified to achieve higher reliability in the performance of electronic circuits.

Figure 4. (PWM) pulse width modulator device

The spectrometer receives the final signal after the laser beam has been pumped through the pump probe system. The characteristics of light across the electromagnetic spectrum may be measured with a spectrometer. In spectroscopic analysis, it is often employed for material identification and the measurement of emission line strengths. The used spectrometer must be fast, with uniform response times between pulses[13]. For this purpose, we used a (Aurora 4000 GE-UV-NIR). spectrometer with 50 µm slit width and CCD silicon detector for spectrum rang 200-1100 nm, which consists of a lens, an optical fiber, and a detector aperture, and which is connected to a computer via a hardware program that displays the results of the analysis of the signal entering the spectrometer and through which the integration time is controlled, a multi-mode optical fiber with a length of 1 meter and a core diameter of 50 µm was used. Fig. 5 show the internal design of spectrometer.

2024, 21(6): 2137-2145
https://doi.org/10.21123/bsj.2023.8383
P-ISSN: 2078-8665 - E-ISSN: 2411-7986

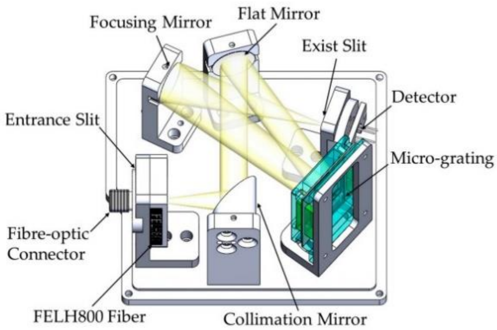


Figure 5. Drawing of the optical layout and assembly of the spectrometer[14]

In a simple way, the mechanisms of the pump-probe system are described in Fig. 6, which shows the integration time of data and the time of the laser pulse when it is interrupted with the delay time, which represents the real time during which the system was checked through recording the photons' quantity passing through it within this time.

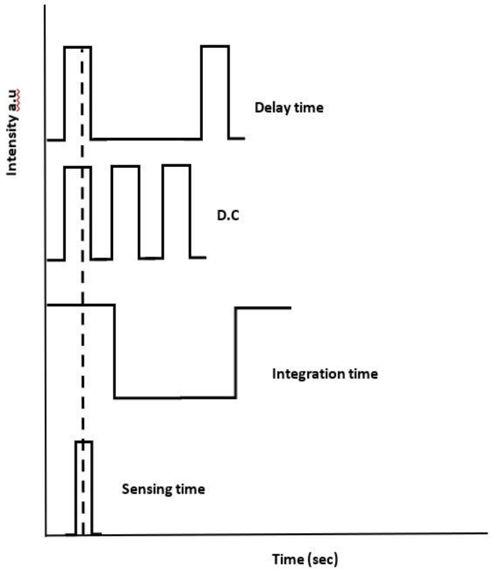


Figure 6. Operation time sequence of the mean element on the pump-probe system

Results and Discussion

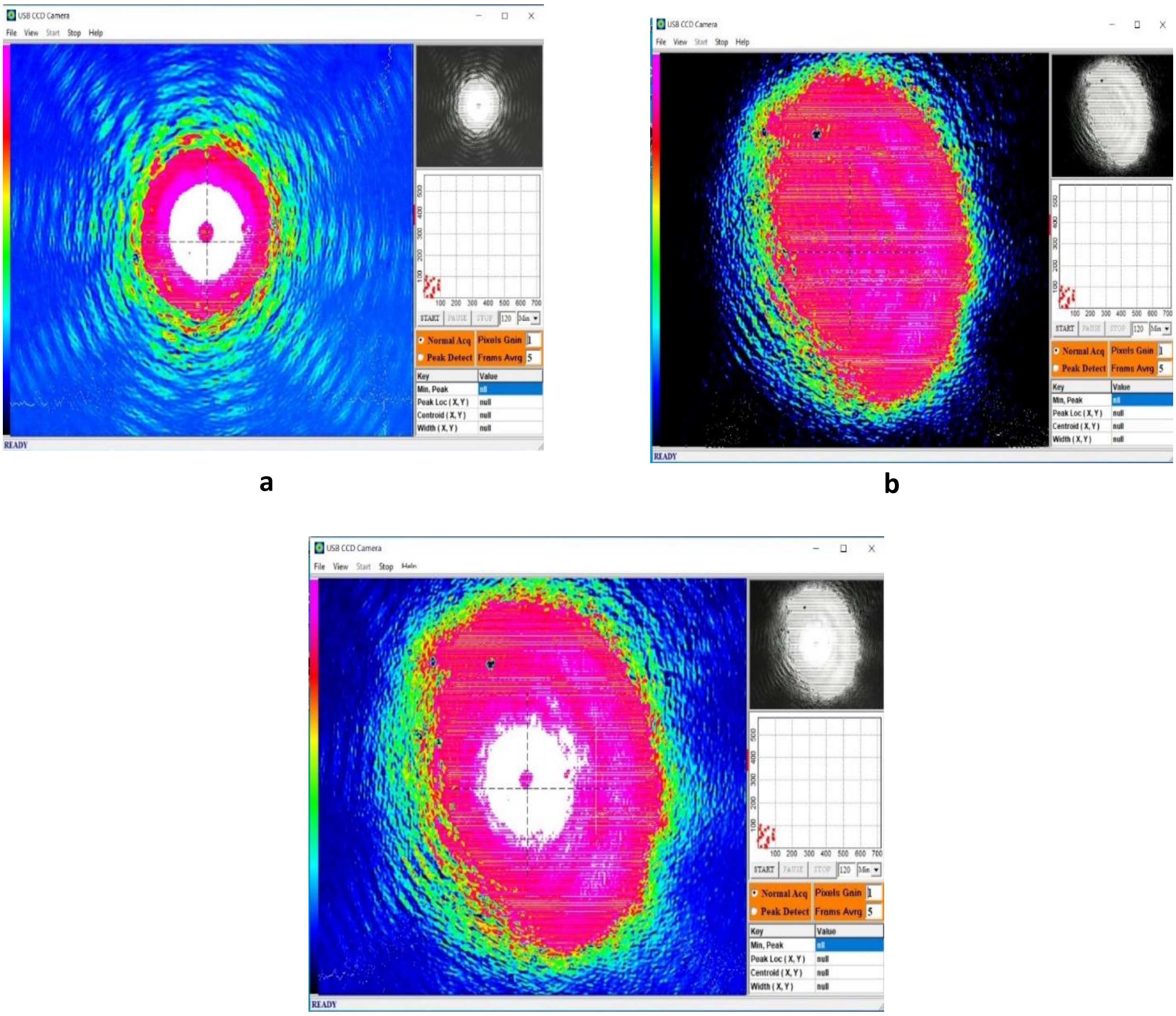
A known frequency Laser beam falls on a beam splitter through it the laser beam is split into two equal beams (50%, 50%) so that the first part **pump** falls on the laser dye directly so it's called **(strongest),** while the second part **probe** passes through a longer optical path than pump can be created by several mirrors so it's called **(weaker)**, thus creating a delay time that can be calculated from Eq. 2[11], Fig. 6 shows a scheme for the work of the system that was designed for the (pump-probe) technique.

The delay time **T** of the laser beam (**Probe**) was calculated through the velocity equation as follows

$$\Delta T = T_2 - T_1$$
$$\Delta T = \frac{d_2}{c} - \frac{d_1}{c}$$
$$\Delta T = \frac{\Delta d\ (m)}{c\ (m/sec)} \qquad 2$$

Where $\mathbf{T_1}$ is the time of pulse in the first path, $\mathbf{T_2}$ **is** the time of pulse in the second path, $\mathbf{d_1}$ first optical path, $\mathbf{d_2}$ second optical path, and **c** speed of light. Also, (the laser beam profile) was seen for pump and prob separately, as well as after overlapping them through the CCD camera (silicon CCD camera for detection range (380-1100) nm with chip dimension (1.2*1.2) cm as shown in Fig. 7.

2024, 21(6): 2137-2145
https://doi.org/10.21123/bsj.2023.8383
P-ISSN: 2078-8665 - E-ISSN: 2411-7986



a b

c

Figure 7. Laser beam profile: A. for bump B. for probe C. after overlap

This system was designed and employed through (**pump - probe**) technique to study the changes in the pulse width (D.C) and the integration time for a range of frequencies with a delay time of (T=12.4 **n** sec) and the following results were found

Fig. 8 shows the relationship between pulse width (D.C.) and integration time. At an integration time of (100, 200) sec, the spectrometer detector's response is equivalent; therefore, pulse width is not dependent on integration time. When the integration time is 50 sec., the detector's response differs from lower values. As shown in Fig. 8, the detector's response to different integration time values is identical at one point where the pulse width is approximately 50% and the detector recorded one intensity for the time. The detector's response is independent of integration time at 50% pulse width, and the intensities were approximately 8000 a.u.

In Fig. 9, the spectrometer detector response is non-existent at a pulse width 10% for all integration time values due to the mismatch between the detector response time and the laser pulse width. When the pulse width was increased to 20%, the detector's response was at 10 sec. and increased with integration time. The maximum recorded value of intensity is at the integration time 10 sec which is the optimal integration time for the system, since after that it continues declining until it records the lowest intensity at the collection time (300 sec) (300 sec). In the case of increasing the pulse width to 50%, we notice that the detector response is compatible with all values of the integration time. Also, we can notice from the Figure. the matching

2024, 21(6): 2137-2145
https://doi.org/10.21123/bsj.2023.8383
P-ISSN: 2078-8665 - E-ISSN: 2411-7986
Baghdad Science Journal

of the detector response at an integration time of 50 sec at an intensity of 8000 au, as well as at an integration time of 225 sec at an intensity of 1600 au for the pulse width of 50% and 20%, respectively.

Figs. 10, 11, and 12 show the frequency-intensity relationship for 10%, 20%, and 50% pulse widths, respectively. We also found that the detector's response to various frequencies increased when the pulse width was increased, and that the detector's response reduced as the frequency was increased for each pulse width. This is due to the disagreement between the frequency values and the pulse width of the source used. And for the integration time of 200 sec., we noticed that the recorded intensity of the detector's response was constant at pulse width 50%, 20% and for all frequencies.

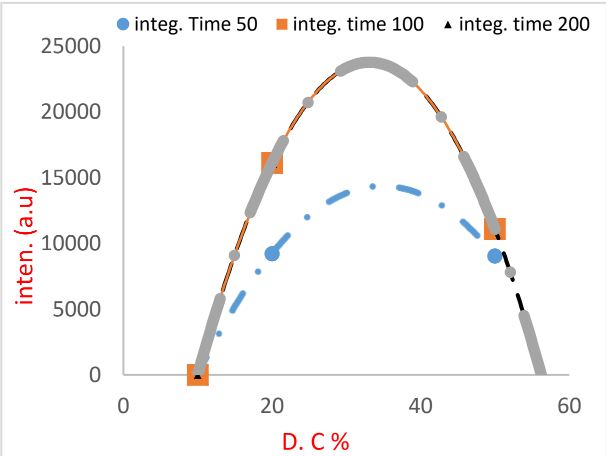


Figure 8. The relation between intensity and pulse width (D.C)

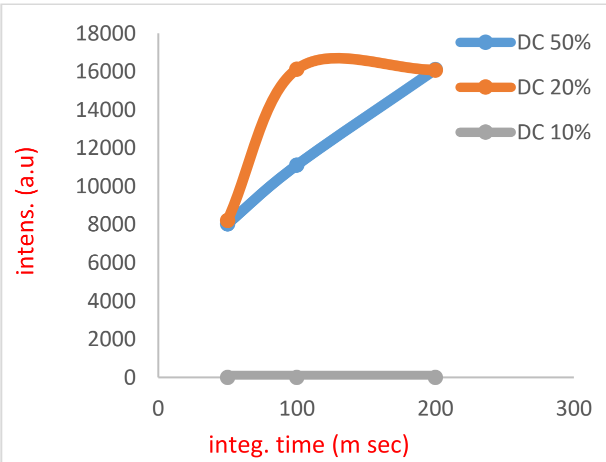


Figure 9. The relation between intensity and integration time

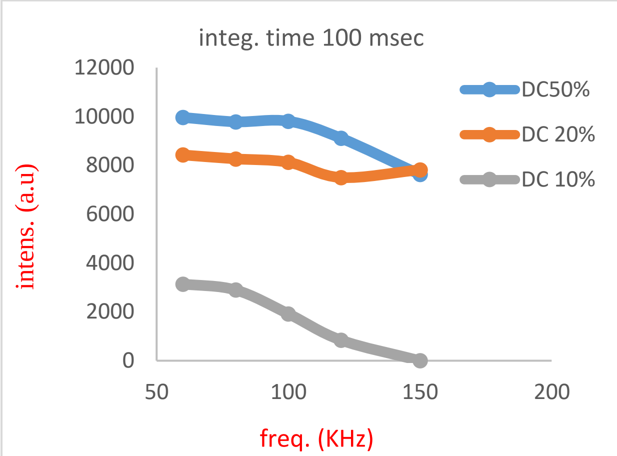


Figure 10. The relation between intensity and frequency for integration time 50msec

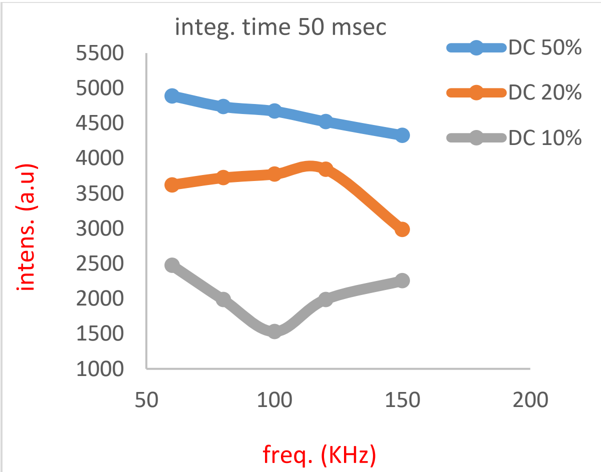


Figure 11. The relation between intensity and frequency for integration time 100msec

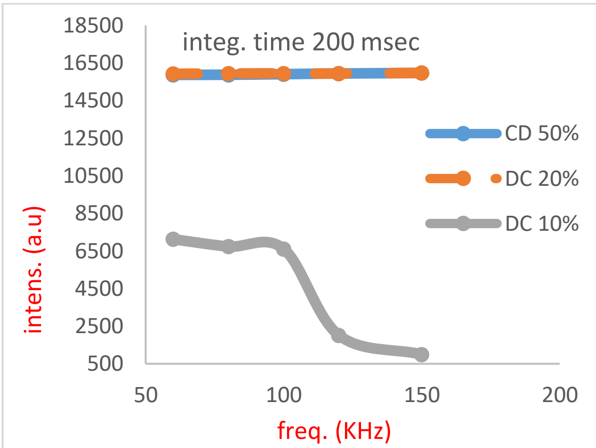


Figure 12. The relation between intensity and frequency for integration time 200msec

Conclusion

Through this research the (Pump-Probe) technique's detector response improves with pump time, the delay time between pump and probe was constant. However, the integration time and laser beam width had a major role in determining the operation of the system at a very specific time. Due to detector-source compatibility, increasing integration time at different frequencies demonstrates the detector's response stability. Controlling the system electrically and optically ensures compatibility in integration times, pumping time (D.C.), and delay time. By changing the delay time between laser pulses, energy level transitions may be monitored. This system can be employed in numerous scientific applications in the fields of spectroscopy, nonlinear optics, and medical applications, as it is characterized by low cost and ease of use, and can be integrated with other devices to obtain various measurements.

Acknowledgment

Authors would be grateful for Mustansiriyah University Iraq- Baghdad for its all support in the present work.

Authors' Declaration

- Conflicts of Interest: None.
- We hereby confirm that all the Figures and Tables in the manuscript are ours. Furthermore, any Figures and images, that are not ours, have been included with the necessary permission for re-publication, which is attached to the manuscript.
- Authors sign on ethical consideration's approval.
- Ethical Clearance: The project was approved by the local ethical committee at University of Mustansiriyah.

Authors' Contribution Statement

This work was carried out under the supervision and follow-up of (S. Kh. R., M. J. Z. and A. H. A.) in addition to conducting laboratory tests, analyzing and discussing the results, S. Kh. R., M. J. Z., And A. H. A. conceived of the presented idea, conceived and planned the experiments. S. Kh. R. carried out the experiments and results. S. Kh. R., M. J. Z., And A. H. A. contributed to the interpretation of the results.

References

1. Hashimoto KY, Kashiwa K, Wu N, Omori T, Yamaguchi M, Takano O, et al. A laser probe based on a Sagnac interferometer with fast mechanical scan for RF surface and bulk acoustic wave devices. IEEE Trans Ultrason Ferroelectr Freq Control. 2011; 58(1): 187-194. https://dx.doi.org/10.1109/TUFFC.2011.1785 .
2. Biesheuvel J, Karr JP, Hilico L, EikemKSE, Ubachs W, Koelemeij JCJ. Probing QED and fundamental constants through laser spectroscopy of vibrational transitions in HD+. Nat Commun. 2016; 7: 10385. https://doi.org/10.1038/ncomms10385 .
3. Komotskii VA, Sokolov YM, Basistyi EV. Depth Measurement of the Periodic Grooved Reflectors of Surface Acoustic Waves Using the Laser Probing. J Commun Technol Electron. 2011; 56(2): 220–225. https://doi.org/10.1134/S1064226910121125 .
4. Harilal SS, Brumfield BE, LaHaye NL, Hartig KC, Phillips MC. Optical spectroscopy of laser-produced plasmas for standoff isotopic Analysis. Appl Phys Rev. 2018; 5: 021301. https://dx.doi.org/10.1063/1.5016053.
5. Giulio C, Cristian M, Larry L, Dario P. Time-resolved methods in biophysics. 4. Broadband pump–probe spectroscopy system with sub-20 fs temporal resolution for the study of energy transfer processes in photosynthesis. Photochem Photobiol Sci. 2007; 6(2): 135–144. https://doi.org/10.1039/B606949E

6. Wu N, Hashimoto Ky, Kashiwa K, Omori T, Yamaguchi M. Study on the frequency dependence of lateral energy leakage in RF BAW device by fast-scanning laser probe system. IEEE Int Ultrason Symp. 2008 Nov. 2-5; Beijing, China. https://dx.doi.org/10.1109/ULTSYM.2008.0023 .
7. Yahya KA. Enhancement of Electron Temperature under Dense Homogenous Plasma by Pulsed Laser Beam. Baghdad Sci J. 2021 Dec; 18(4): 1344. https://doi.org/10.21123/bsj.2021.18.4.1344 .
8. Emre K, Gokhan G. A comparison of fuzzy logic and PID controller for a single-axis solar tracking system. Renewables Wind Water and Solar. 2016 3(1): 7. https://dx.doi.org/10.1186/s40807-016-0023-7
9. Naif HA, Saeed AA, Al-Kadhemy MFH. Spectral Behaviour of the low concentrations of Coumarin 334 with Broadband Cavity Enhanced Absorption Spectroscopy. Baghdad Sci J. 2022 Apr; 19(2): 0438. https://dx.doi.org/10.21123/bsj.2022.19.2.0438
10. Raied KJ, Russul KR. Studying the Effect of Laser Pump Pulse Energy and Delay Time on conversion Efficiency of KTP. Iraqi J Sci. 2020; 61(2): 351-357. https://dx.doi.org/10.24996/ijs.2020.61.2.13.
11. Dong PT, Cheng JX. Pump-Probe Microscopy Theory Instrumentation and Applications. Spectroscopy. 2017; 32(4): 24-36. https://sites.bu.edu/cheng-group/files/2017/10/8-Pu-Ting-Dong-Pump-probe-review-in-Spectroscopy.pdf
12. . Anchi Y, Xiong Y, Dan I, Wenxiang C, Paul MC, Two-color pump-probe laser spectroscopy instrument with picosecond time-resolved electronic delay and extended scan range. Rev Sci Instrum. 2005; 76: 114301. https://dx.doi.org/10.1063/1.2126808.
13. Abramczyk H. Introduction to Laser Spectroscopy. Elsevier. 1st Edition, 2005.
14. Liangkun H, Quan W, Jian H, Fan Y, Hongjie L, Zhiyu W. Miniature Broadband NIR Spectrometer Based on FR4 Electromagnetic Scanning Micro-Grating. Micromachines. 2020; 11: 393. https://dx.doi.org/10.3390/mi11040393.

تصميم وتطوير نظام بصري يعمل بتقنية المجس الليزري للدراسة الطيفية

سعد خالد راهي[1]، مؤيد جبار زوري[1]، علاء حسين علي[2]

[1]قسم الفيزياء، كلية العلوم، الجامعة المستنصرية، بغداد، العراق.
[2]وزارة العلوم والتكنلوجيا، بغداد، العراق.

الخلاصة

في هذا البحث، تم تصميم نظام بصري باستخدام تقنية المجس الليزري لدراسة تأثير تغيير قيم التردد وزمن التجميع وزمن التأخير وعرض نبضة الليزر لطاقة نبضة الضخ على مادة صبغة ليزرية عشوائية ذات معامل امتصاص حوالي 650 نانومتر. وجد أن تقليل زمن الضخ إلى 10٪ من زمن النبضة غير مناسب لتقنية المجس الليزري بسبب عدم التناغم بين زمن الضخ واستجابة الكاشف للمصدر المستخدم. كذلك، عند زيادة زمن الضخ إلى 20٪ من زمن النبضة، تكون أستجابية الكاشف عالية قدر الإمكان عند زمن التجميع 150 مايكرو ثانية وبالتالي لوحظ أن أستجابية الكاشف أكثر ملاءمة للعمل في هذه التقنية حيث يزداد زمن الضخ، وكذلك كفاءة الكاشف، والتي تزداد بشكل ملحوظ مع زيادة زمن الضخ، وكذلك زمن التجميع لنفس زمن التأخير، ويكون أكثر ثباتًا عند 20٪ من زمن الضخ وزمن التجميع 150 ما يكرو ثانية في زمن تأخير يبلغ 12.4 نانو ثانية.

الكلمات المفتاحية: زمن التأخير، زمن التجميع، كاشف بصري، عرض النبضة، تقنية المجس الليزري.