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GRADUATE UNIVERSITY SCIENCE AND TECHNOLOGY

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RESEARCH AND DEVELOPMENT OF AN ULTRAWIDE-BAND ULTRAVIOLET LASER SYSTEM USING CE:LICAF CRYSTALS FOR AIR POLLUTANT MONITORING

Major: Optical

Code: 944 01 10

SUMMARY OF SCIENCE MATERIAL DOCTORAL THESIS

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The thesis was completed at:

Department Physics, Graduate University Science and Technology - Vietnam Academy of Science and Technology

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Reviewer 2:
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INTRODUCTION

1. The urgency of thesis

Introduced in 1960, lasers have rapidly developed in terms of resonant cavity configuration, pumping techniques, and active media, significantly expanding the wavelength range, power, and emission modes. Thanks to outstanding characteristics such as coherence, directionality, monochromaticity, and high power, laser radiation has become a crucial light source in modern optics. Laser sources in general, and broadband lasers in particular, are widely used in scientific research and everyday life, directly driving the formation and development of many modern scientific and technological fields such as metrology, biomedical science, and environmental research.

Atmospheric gases contain many types of pollutants with characteristic absorption spectra in the ultraviolet (UV) wavelength region, such as O_3 (200–400 nm), SO_2 (280–300 nm), NO_2 (200–400 nm), HCHO (250–400 nm), C_6H_6 (220–260 nm), etc. Many highly accurate methods for detecting and quantifying these pollutants have been researched and effectively applied. Among them, DOAS stands out with its advantages: relatively simple optical system configuration, reasonable deployment cost, the ability to simultaneously measure multiple gas components, and particular suitability for remote measurements. Using a wide-spectrum UV laser as the excitation source in DOAS systems expands the spectral range under investigation and increases the ability to simultaneously detect multiple gases. Therefore, the development of ultra-broadband UV laser sources for use as excitation sources in DOAS systems studying air pollutants with absorption spectra in the UV region is necessary.

From the above analysis, it is clear that to further expand the application of UV laser sources in research and practical applications, the development of ultra-broadband, high-energy UV laser sources is essential. Based on the aforementioned scientific and practical requirements, I have chosen this research topic **"Research and development of an ultrawide-band ultraviolet laser system using Ce:LiCAF crystals for air pollutant monitoring"** as the main research direction for my thesis.

2. The purpose of the thesis:

This study investigates the kinetics of emission and amplification of modeless ultraviolet laser pulses using Ce:LiCAF crystals. Based on theoretical research results, a system for emitting and amplifying modeless ultraviolet laser pulses using Ce:LiCAF crystals is developed.

The application of modeless Ce:LiCAF ultraviolet laser sources in the study of SO_2 and O_3 air pollution is also discussed.

3. The main research contents of the thesis

This study investigates the theoretical and experimental development of a super-wideband, modeless ultraviolet pulsed signal laser system and a super-wideband, modeless ultraviolet laser pulse amplifier system using Ce:LiCAF crystals. The initial research focuses on the application of modeless ultraviolet laser sources with wavelengths of 275 nm - 330 nm in the study of SO_2 and O_3 air pollutants.

CHAPTER 1: OVERVIEW OF ULTRAWIDE-BAND ULTRAVIOLET LASERS AND THEIR APPLICATIONS

1.1. Overview of ultrawide-band ultraviolet laser emission

1.1.1. Frequency-Variable Lasers

Generating broad-spectrum UV radiation using IR or VIS laser sources is a process that exploits the high-order nonlinearity of materials when interacting with strong electromagnetic fields, typically femtosecond pulses. Frequency multiplication requires control over the phase alignment conditions between the generated nonlinear wave and its corresponding nonlinear polarization wave within the crystal, as well as the characteristics of the nonlinear medium.



Fig 1.1. Diagram illustrating the frequency conversion process

1.1.2. Supercontinuous laser

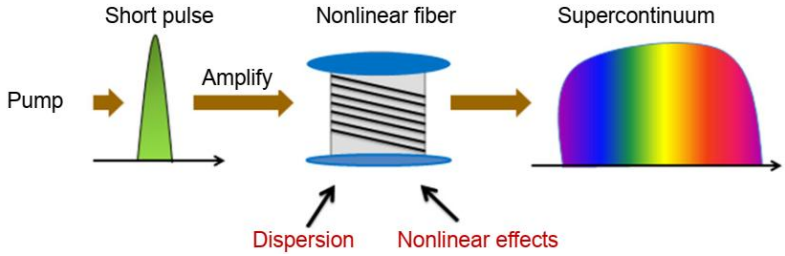


Fig 1.3. Diagram illustrating the process of supercontinuous transmission

Supercontinuous emission is a complex nonlinear optical phenomenon. The formation of SC spectra in the UV region results from a series of interactions between high-power pulsed lasers, typically femtosecond pulses, and a nonlinear medium including:

phase automodulation; group dispersion; and soliton dynamics and soliton splitting processes. A schematic representation of the supercontinuous emission process is shown in Fig 1.3.

1.1.3. Modeless lasser

A modeless laser is a type of laser designed to avoid forming distinct resonant modes in the BCH or to significantly attenuate their existence. Modeless lasers have a spectrum that is not split into discrete longitudinal modes. The modes are obscured or unstable, resulting in a continuous and very broad emission spectrum. The modeless laser spectrum is shown in Fig 1.5c.

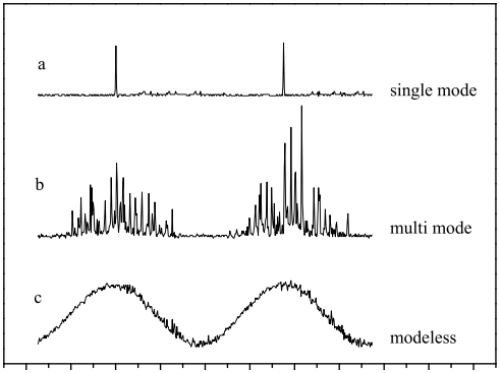


Fig 1.5. a) Spectrum laser single mode, b) Spectrum laser multi mode, c) Spectrum of laser modeless

1.2. Air pollution and methods for studying air pollution

1.2.1. Overview of air pollution

Air pollution is one of the most serious environmental problems today. Although it accounts for only a very small percentage of the atmosphere, it negatively impacts human health, ecosystems, and the global climate.

1.2.1.1. Sulfur dioxide (SO_2)

1.2.1.2. Ozone (O_3)

1.2.1.3. Some other polluting gases

1.2.2. Some methods for studying air pollution

1.2.2.1. Laser-Induced Fluorescence

1.2.2.2. Cavity Ring-Down Spectroscopy

1.2.2.3. Differential Optical Absorption Spectroscopy

1.3. Applications of ultraviolet lasers in air pollution research

In 2019, Jan-Marcus Nasse and colleagues published a study on a high-precision, stable, and long-term autonomous differential long-distance absorption spectroscopy (LP-DOAS) system for atmospheric monitoring. The system measures atmospheric emissions over optical trajectories from a few hundred meters to a few kms.

1.4. Overview of Cerium Fluoride Solid-State UV Lasers

1.4.1. Cerium Fluoride medium

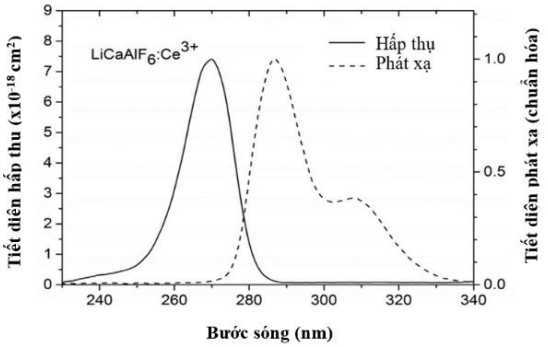


Fig 1.10. Absorption and emission spectra of Ce:LiCAF

Among Ce-Fluoride media, Ce:LiCAF is an efficient ultraviolet laser emission and amplification medium with many outstanding advantages. The medium strongly absorbs at a wavelength

of 266 nm (Fig 1.10). It has a large emission cross-section and a wide emission spectrum from 275 nm to 325 nm. The fluorescence lifetime of the excited Ce^{3+} ion is 25 ns to 30 ns, suitable for developing multi-pass-through laser amplification systems. Furthermore, the medium has a high saturation energy density of approximately 115 mJ/cm^2 and a high threshold of 5 J/cm^2.

1.4.2. Laser Ce:LiCAF

Among the Ce^{3+} ion-doped fluoride laser media, Ce:LiCAF crystals have been shown to be an effective direct UV radiation emission medium. Ce:LiCAF crystals have been selected by many research groups worldwide for the development of wavelength-tuned narrowband UV lasers, short-pulsed lasers, and high-power lasers. Recent research on Ce:LiCAF lasers has primarily focused on wavelength-tuned narrowband lasers, short-pulsed lasers, and high-power lasers through amplification. However, this medium shows that Ce:LiCAF also has enormous potential for developing ultra-broadband UV laser sources. Therefore, research and development of ultrawide-band UV laser sources using Ce:LiCAF crystals is not only of scientific significance but also opens up new application avenues for this laser medium in absorption spectroscopy, lidar, and environmental monitoring.

Summary of chapter 1: In Chapter 1, through an analysis of worldwide research results on modeless laser emission and ultra-wide emission spectra, the thesis identified the following key issues: An overview of the development and application directions of ultrawide-band UV lasers was presented. The development trend of modeless lasers was analyzed, showing that modeless laser configurations are a

suitable approach to generating UV radiation with ultra-broadband spectra. Important characteristics of Ce:LiCAF crystals indicate that this is a suitable active medium for the development of ultra-broadband ultraviolet laser systems. From the overview results, it can be affirmed that the research and development of ultrawide-band modeless Ce:LiCAF UV lasers combined with DOAS techniques is a research direction with high scientific and practical significance.

CHAPTER 2: RESEARCH AND DEVELOPMENT OF ULTRAWIDE- BAND MODELESS ULTRAVIOLET LASER SYSTEMS USING CRYSTALS Ce:LiCAF

2.1. Theoretical model for the spectral dynamics of modeless ultraviolet laser

2.1.1. Schematic design of a modeless ultrawide-band Ce:LiCAF laser system

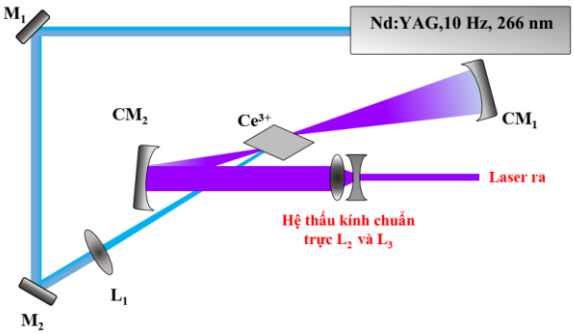


Fig 2.1. Schematic design of a modeless ultrawide-band Ce:LiCAF laser system

2.1.2. The rate equations describing the kinetics of modeless laser emission

2.1.3.

$$dN_2(z,t)/dt = \eta_p\,\sigma_{ap}(\lambda_p)\,\Phi_p(z,t)\,N_1(z,t) - N_2(z,t)/\tau_f$$
$$-\Gamma_s \sum_i \frac{I_i(z,t)}{h\nu_i}[\sigma_e(\lambda_i)N_2(z,t) + \sigma_a(\lambda_i)N_1(z,t)]$$

$$\partial I_i(z,t)/\,\partial z = g_i(z)\,I_i(z,t) + S_{sp,i}(z,t) - \alpha(\lambda_i)\,I_i(z,t)$$
$$g_i(z) = \Gamma_s[\sigma_e(\lambda_i)\,N_2(z) - \sigma_a(\lambda_i)\,N_1(z)]$$

2.1.4. Parameters used in simulating the dynamics of modeless laser spectra

To study the emission kinetics of a modeless Ce:LiCAF ultraviolet laser, the parameters used in the simulation are presented in Table 2.1.

2.1.5. Nonmode ultraviolet laser kinetics using Ce:LiCAF crystals

The spectral dynamics of the ultrawide-band Ce:LiCAF UV laser in a modeless laser configuration were studied by solving the system of equations (2.1) - (2.3) using MATLAB software. With a pump laser energy of 10 mJ at a wavelength of 266 nm and a pulse width of 7 ns, the emission spectrum of the Ce:LiCAF UV laser is shown in Fig 2.2.

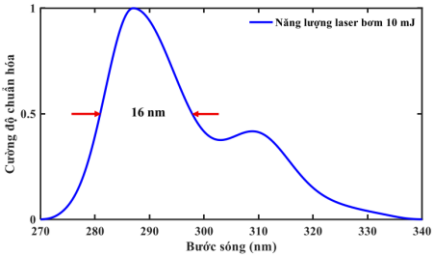


Fig 2.2. Ce:LiCAF modeless UV laser emission spectrum

The dependence of the output laser energy on the pump laser

energy has been simulated and detailed. The survey results indicate that the output laser energy increases linearly with increasing pump energy in the surveyed range from 4 mJ to 14 mJ.

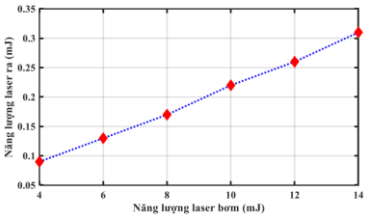


Fig 2.4. Effect of pump laser energy on output laser energy

2.2. Development of a superwide-band modeless Ce:LiCAF laser experimental system

2.2.1. Components and equipment used

To construct the ultrawide-spectrum Ce:LiCAF modeless laser system, this thesis utilizes several components and equipment from the Institute of Physics.

2.2.2. Experimental system ultrawide-band modeless Ce:LiCAF laser

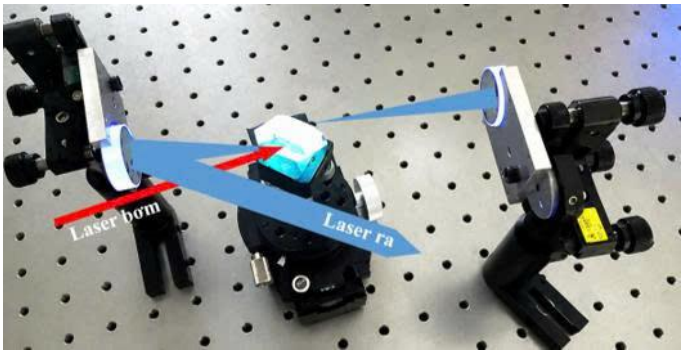


Fig 2.5. Experimental system ultrawide-band modeless Ce:LiCAF laser

2.2.2.1. Spectral characteristics of modeless Ce:LiCAF ultraviolet laser

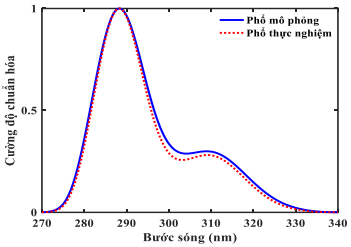


Fig 2.6. Ce:LiCAF modeless ultraviolet laser spectrum

2.2.2.2. Pulse characteristics of modeless Ce:LiCAF ultraviolet laser

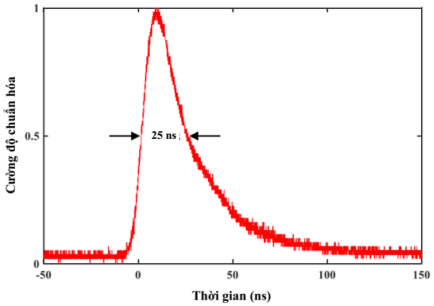


Fig 2.7. Ce:LiCAF ultraviolet laser pulses modeless

2.2.2.3. Divergence of modeless Ce:LiCAF ultraviolet laser beam

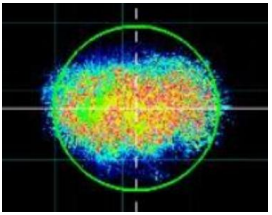


Fig 2.8. Cross-sectional image of the laser beam at position Z = 10 cm obtained from CCD

Measurement results show that the average total FWHM divergence of the signal laser beam along the X-axis is 4.07 ± 0.30 mrad, and along the Y-axis is 2.50 ± 0.29 mrad. Thus, the output laser beam has an elliptical shape with significantly greater X-axis divergence than Y-axis divergence.

2.2.2.4. Energy stability of modeless Ce:LiCAF UV laser beam

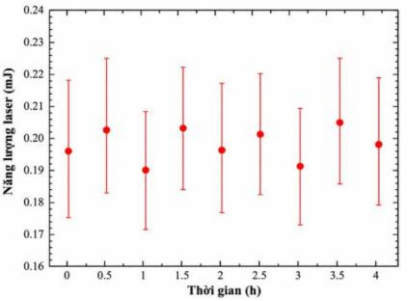


Fig 2.9. Evaluating the stability of modeless laser energy

2.2.2.5. Effect of pump laser energy on output laser energy

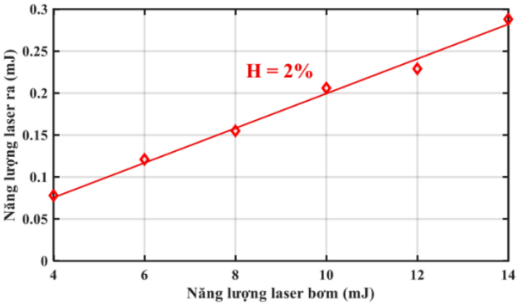


Fig 2.10. The dependence of the output laser energy on the pump laser energy

Summary of chapter 2: In Chapter 2, the emission kinetics for ultra-wideband Ce:LiCAF UV lasers using modeless configurations were investigated. A successful development of a

Ce:LiCAF UV laser system using a modeless configuration achieved a laser efficiency of 2%, with a maximum laser energy of 0.3 mJ at a pump energy of 14 mJ.

CHAPTER 3: RESEARCH AND DEVELOPMENT OF Ce:LiCAF MODELESS ULTRAWIDE-BAND ULTRAVIOLET LASER AMPLIFIERS

3.1. Amplification kinetics of ultrawide-band modeless ultraviolet laser pulses using Ce:LiCAF crystals

3.1.1. Design diagram of a modeless ultraviolet laser pulse amplifier system

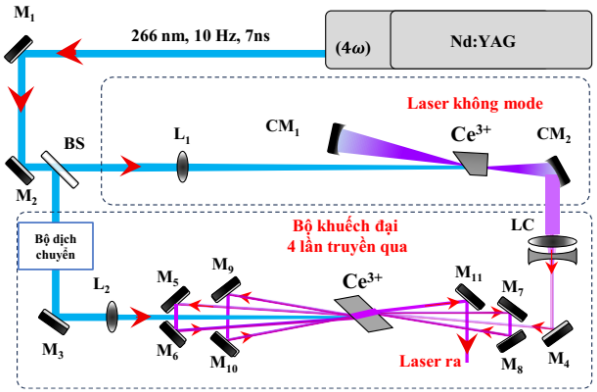


Fig 3.1. Schematic design of system amplifier ultrawide-band ultraviolet laser pulses with 4-pass through a Ce:LiCAF crystal

3.1.2. Theoretical model for amplification of modeless ultraviolet laser pulses

$$F_{\text{out}}^{(n)}(\lambda_i) = F_{\text{sat}}(\lambda_i)\ln\left[1+\exp\left(G_0(\lambda_i)\right)\left(\exp\left[F_{\text{in}}^{(n)}(\lambda_i)/F_{\text{sat}}(\lambda_i)\right]-1\right)\right]$$

$$G_0(\lambda_i) = \Gamma_s\sigma_e(\lambda_i)N_{2,0}^{(n)}l - \alpha(\lambda_i)l$$

$$F_{sat}(\lambda_i) = (hc/\lambda_i)/\sigma_e(\lambda_i)\,.$$

3.1.3. Parameters used in the simulation

The parameters used in the simulation of amplifier UV laser pulses using Ce:LiCAF crystals were obtained from existing components and equipment at the Photonics Laboratory - Institute of Physics and are presented in detail in Table 3.1.

3.1.4. Amplification kinetics of ultrawide-band ultraviolet laser pulses

3.1.4.1. Effect of pump laser energy on output laser energy

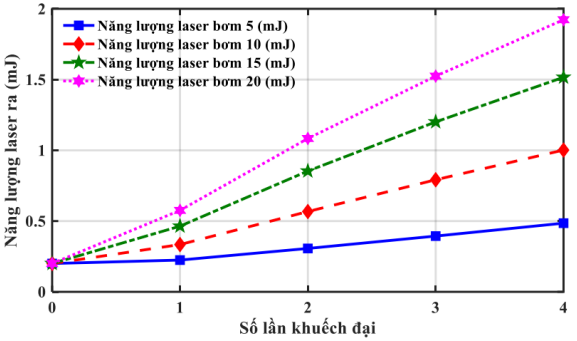


Fig 3.3. Effect of pump energy on laser energy after amplification

3.1.4.2. Effect of signal laser energy on output laser energy

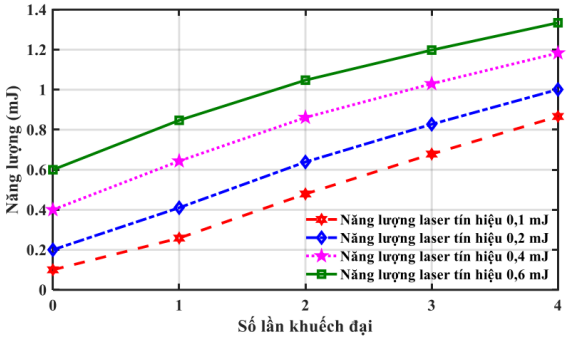


Fig 3.4. Effect of signal laser energy on amplified laser energy

3.1.4.3. Effect of signal laser spectral width on output laser energy

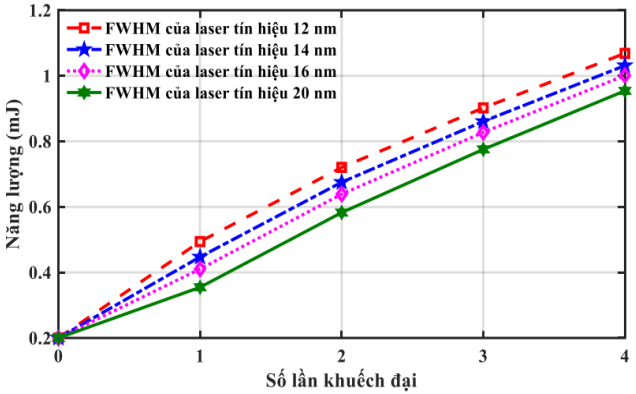


Fig 3.5. Effect of the signal laser spectral width on the amplified laser energy

3.1.4.4. Narrowing the spectrum during amplification

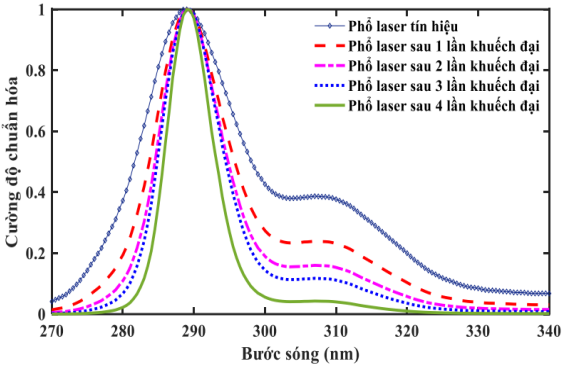


Fig 3.6. Laser spectrum after each amplification stage

3.2. Ce:LiCAF 4-pass Ultrawide-band Modeless Ultraviolet Laser Amplifier System

3.2.1. Experimental system amplifier Ce:LiCAF modeless ultraviolet laser

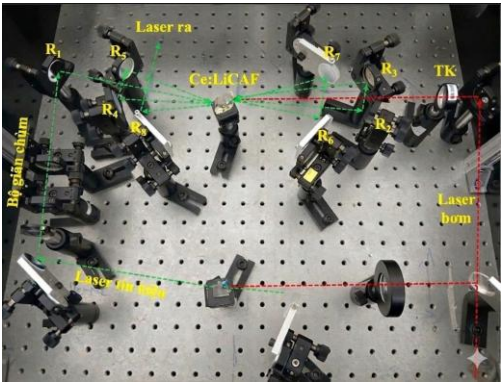


Fig 3.7. Ultrawide-band UV laser pulse amplifier system 4-pass using Ce:LiCAF crystals

3.2.2. Energy characteristics during the amplification process

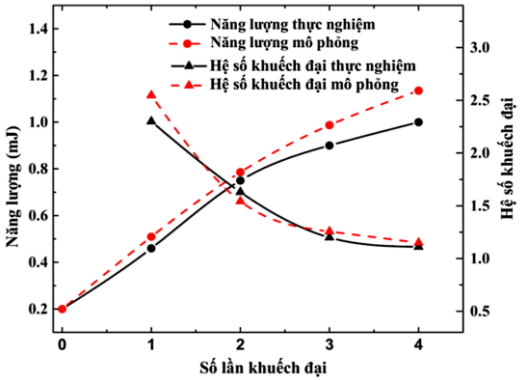


Fig 3.8. The change in laser energy and amplification factor after each transmission

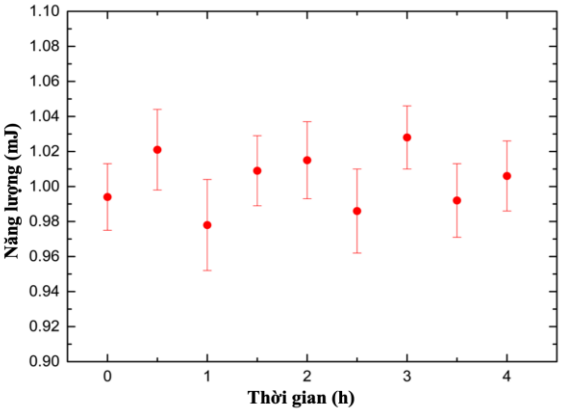


Fig 3.9. Evaluating the stability of the laser energy after amplification

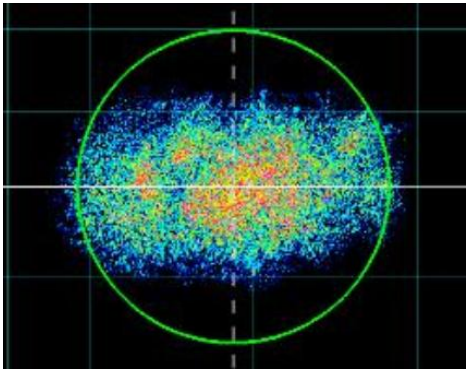


Fig 3.10. Cross-sectional image of the laser beam obtained from a CCD camera

3.2.3. Spectral characteristics during amplification

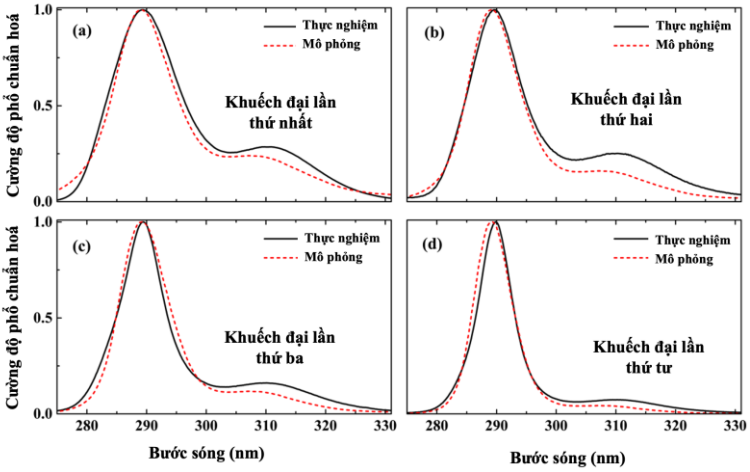


Fig 3.11. Spectral simulation results (red dashed line) and experimental results (black solid line) from the ultrawide-band ultraviolet laser pulse amplifier system

3.2.4. Time characteristics of the laser pulse during amplification

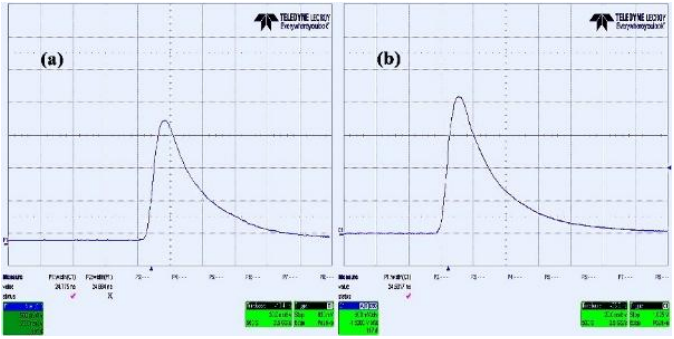


Fig 3.12. (a) Signal laser pulse and (b) Laser pulse after 4 amplifications

Summary of chapter 3: The results of the research on developing a signal amplification system for a modeless, ultra-wideband emission Ce:LiCAF ultraviolet laser have shown:

A multi-wavelength amplification model based on the extended Frantz–Nodvik equation was constructed to describe the amplification kinetics of an ultra-wideband emission modeless UV laser in a Ce:LiCAF crystal. Through simulation, the spectral amplification kinetics of the ultra-wideband UV laser were quantitatively clarified. The results show that the laser pulse energy increases significantly with the number of passes through the Ce:LiCAF crystal, from approximately 0.2 mJ to nearly 1 mJ after 4-passes. The phenomenon of spectral narrowing during amplification was investigated and analyzed. Simulation results show that the FWHM spectral width decreases from approximately 16 nm to approximately 7 nm after four passes due to the spectral components near the amplification peak being amplified more strongly than those at the spectral edge. Experimental results show good agreement with the simulation. The laser output energy reached nearly 1 mJ after four passes, and the laser spectrum showed a similar narrowing trend as calculated, with the spectral peak remaining stable in the region of approximately 288–290 nm.

CHAPTER 4: APPLICATION OF ULTRAWIDE-BAND MODELESS ULTRAVIOLET LASER SOURCES IN AIR POLLUTION RESEARCH

4.1. Experimental System and Data Processing Methods for DOAS

In this chapter, by developing a DOAS system using a modeless ultra-wideband Ce:LiCAF UV laser as the excitation source, the pollutants SO_2 and O_3 will be studied.

4.1.1. Experimental system for measuring air pollutant concentrations

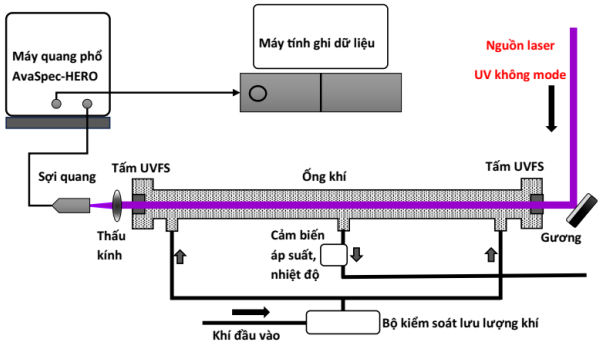


Fig 4.1. Overall diagram of the experimental system for measuring gas absorption using modeless ultrawide-band UV laser

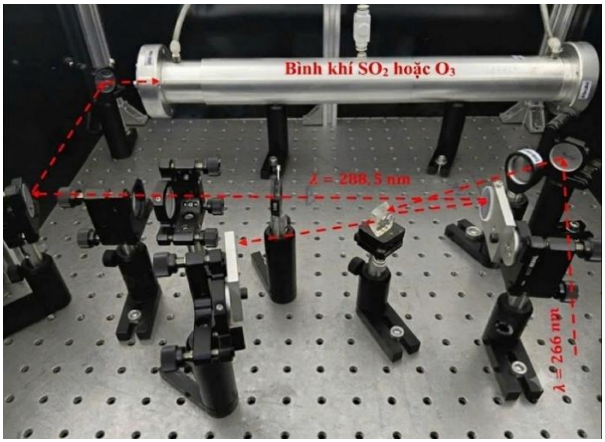


Fig 4.2. Photograph of an experimental setup for measuring air pollution in a laboratory

4.1.2. DOAS Method and Data Processing Procedure

Essentially, the DOAS method is based on the Beer-Lambert law, which describes the attenuation of light intensity as it passes through an absorbing medium. The data analysis process is built on the theoretical basis of the DOAS method and the QDOAS software developed by BIRA-IASB.

4.2. Calibration of the measurement system using SO_2 standard gas

4.2.1. *Analysis of standard SO2 gas measurement data*

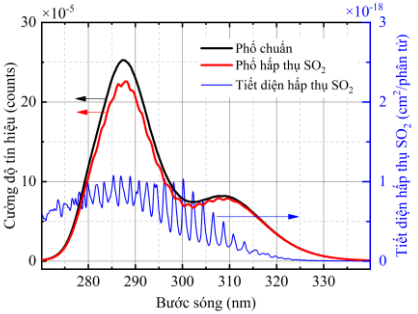


Fig 4.6. Standard spectra, absorption spectra and standard absorption cross-section spectra of SO_2

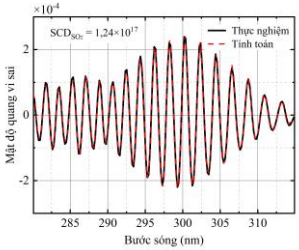


Fig 4.7. Results of differential spectrum matching of standard SO_2 gas using QDOAS software

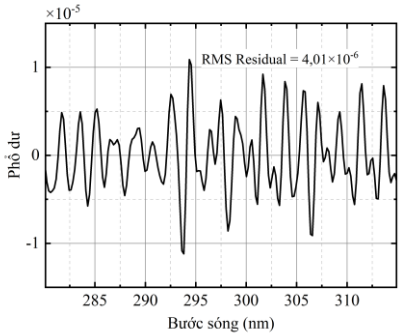


Fig 4.8. Post-match residual spectrum for SO_2 standard gas

4.2.2. Evaluating the accuracy and reliability of the measurement system

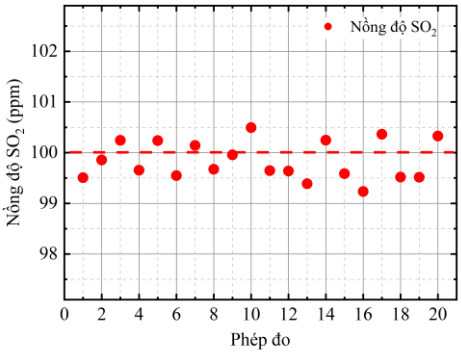


Fig 4.9. Statistical results of SO_2 concentrations recovered from 20 repeated measurements with a 100 ppm standard gas

4.3. Survey of O_3 gas concentration

After the measurement system has been calibrated with standard SO_2 gas and the recovery results match the nominal concentration, the next step is to investigate the applicability of the

experimental system to O_3 gas produced from the gas generator.

4.3.1. Analysis of O_3 gas measurement data

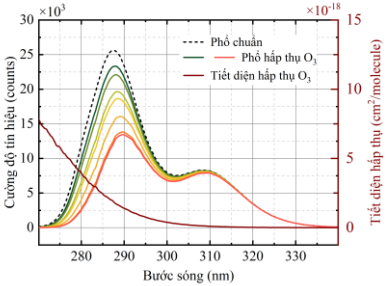


Fig 4.10. Standard spectra, absorption spectra and standard absorption cross-section spectra of O_3

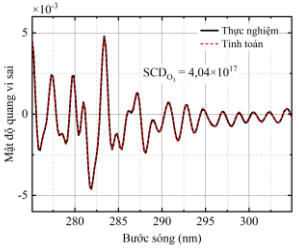


Fig 4.11. Differential spectral matching results for O_3 gas using QDOAS software

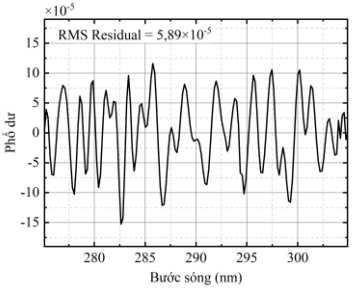


Fig 4.12. Post-match residual spectrum for O_3 gas

4.3.2. Results of the O_3 gas concentration survey

Order	Time (m)	*C* (ppm)	±Δc (ppm)	SNR	RMS
1	Bắt đầu	327,75	± 2,45	133,8	$5,89\times10^{-5}$
2	~5	306,99	± 2,64	116,3	$5,79\times10^{-5}$
3	~20	225,70	± 2,82	82,1	$5,57\times10^{-5}$
4	~35	149,42	± 2,92	51,2	$5,37\times10^{-5}$
5	~50	123,99	± 2,89	42,9	$5,33\times10^{-5}$
6	~65	68,04	± 2,85	23,9	$5,26\times10^{-5}$
7	~80	52,80	± 2,83	18,7	$5,22\times10^{-5}$
8	~90	36,68	± 2,79	13,1	$5,17\times10^{-5}$
9	~100	13,19	± 2,80	4,7	$5,13\times10^{-5}$

Summary of chapter 4: In this chapter, a non-mode ultra-wideband UV laser source was used to construct an experimental system for measuring gas absorption in the laboratory using the DOAS method. Calibration with the standard SO_2 gas showed that the system recovered a concentration of approximately 99.7 ± 1 ppm, consistent with the nominal concentration of 100 ± 2 ppm of the standard gas. For O_3 gas from the gas generator, the system not only detected and recovered the O_3 concentration but also monitored the decrease in concentration over time. The O_3 concentration decreased from approximately 327.75 ppm at the beginning to 13.19 ppm after about 100 minutes.

GENERAL CONCLUSION:

This thesis successfully conducted comprehensive research, from theoretical foundations and simulations to experimental development of a modeless ultrawide-band UV Ce:LiCAF laser source, studying amplification kinetics, and applying this laser source to measure SO_2 and O_3 pollutant concentrations using the DOAS method. The results achieved have contributed to clarifying the emission and amplification characteristics of ultrawide-band UV lasers, and demonstrated their potential applications in environmental monitoring. The research results confirm that the modeless ultrawide-band UV Ce:LiCAF laser source is a promising light source for optical measurement systems in the UV region. The thesis not only contributes new results on the emission and amplification kinetics of ultrawide-band UV lasers based on Ce:LiCAF crystals but also successfully develops a laser source system and initially demonstrates its effectiveness in measuring pollutant concentrations using the DOAS method.

The results of this thesis provide a scientific and experimental basis for the further development of ultrawide-band UV laser sources with higher power and stability, and open up prospects for applications in optical environmental monitoring systems, remote measurement of air pollution, and high-sensitivity UV spectroscopy methods in the future.

LIST OF PUBLICATIONS

ISI Journal

1. **Quan Minh Le**, Tu Xuan Nguyen, Diep Van Nguyen, Kieu Anh Thi Doan, Bich Thi Vu, Anh Le Ngoc, Minh Hong Pham, Nobuhiko Sarukura, Hung Nguyen Dai, *Development of an ultrawide-band modeless Ce:LiCAF ultraviolet laser for SO_2 detection using differential optical absorption spectroscopy*. Vol. **594**, 2025, **Optics Communications**, pp. 132323.

National magazine

1. Nguyen Xuan Tu, Nguyen Van Diep, **Le Minh Quan**, D.T. Kieu Anh, Le Ngoc Anh, Nobuhiko Sarukura, and Pham Hong Minh, *Numerical and experimental study of an ultrabroadband modeless Ce:LiCAF ultraviolet laser: multiwavelength dynamics and power amplification.* Vol. **36**, 2026, **Communications in Physics**, pp. 205 - 218.

Conference Proceedings

1. **Lê Minh Quân**, Nguyễn Văn Điệp, Nguyễn Xuân Tú, Phạm Đức Tú, Trần Đức Minh, Phạm Văn Dương, Phùng Việt Tiệp, Nguyễn Thị Nhung, Đặng Hồng Lưu, Nguyễn Thị Khánh Vân, Đoàn Hoài Sơn, Vũ Thị Bích, Phạm Hồng Minh, *Nghiên cứu động học laser tử ngoại phổ rộng Ce:LiCAF sử dụng buồng cộng hưởng có hệ số phản xạ gương thay đổi theo bước sóng*, **Những tiến bộ trong Quang học, Quang tử, Quang phổ và Ứng dụng XIII. ISBN: 978-604-357-381-7**, 2025, pp. 498-502.
2. **Lê Minh Quân**, Nguyễn Văn Điệp, Nguyễn Xuân Tú, Phạm Văn Dương, Lê Hồng Việt, Lê Ngọc Anh, Phạm Đức Tú, Vũ Dương, Vũ Thị Bích, Phạm Hồng Minh. *Nghiên cứu và phát triển hệ laser tử ngoại phổ siêu rộng cho ứng dụng trong nghiên cứu khí ô nhiễm*, **Tuyển tập báo cáo Hội nghị khoa học kỷ niệm 50 năm thành lập Viện Hàn lâm**. **ISBN: 978-604-357-359-6**, 2025, pp. 250 - 256**.**
3. **Le Minh Quan**, Nguyen Van Diep, Nguyen Xuan Tu, Tran Duc Minh, Doan Thi Kieu Anh, Pham Duc Tu, Pham Hong Minh., *Characterization of ultraviolet all-solid-state modeless Ce:LiCAF laser*, **CASEAN-9**. **ISBN: 978-604-357-473-9**, 2025, pp. 359 - 363**.**
4. Diep Van Nguyen, Tu Xuan Nguyen, **Le Minh Quan**, Hieu Minh Do, Kieu Anh Thi Doan, Nguyen Thi Nhung, Le Hong Viet, Dang Thi Vui & Minh Hong Pham. *Spectral dynamics of ultra-broadband laser generation and amplification using Ce:LiCAF crystal*, **CASEAN-9**. **ISBN: 978-604-357-473-9**, 2025, pp. 258 – 292.