

**Near-infrared Femtosecond Laser Sources Based on
Alexandrite and Cr:LiSAF Gain Media**

by

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Abstract

Alexandrite ($\text{Cr}^{3+}:\text{BeAl}_2\text{O}_4$) and Cr:LiSAF ($\text{Cr}^{3+}:\text{LiSrAlF}_6$) are promising near-infrared gain media. They have tuning ranges extending from 700 nm to 820 nm and from 780 nm to 1110 nm, broad enough to support sub-10 fs pulse generation. They can be directly pumped with low-cost red diodes. Furthermore, since the emission cross section-fluorescence lifetime products of these media are greater than that of Ti:sapphire ($\text{Ti}^{3+}:\text{Al}_2\text{O}_3$), they have low laser thresholds. Due to their high quantum efficiency and high figure of merit, these gain media can be utilised as low-cost, robust, compact and efficient alternatives to Ti:sapphire lasers near 800 nm. In particular, Alexandrite has superior thermo-mechanical properties which make it suitable for the development of high power laser applications. Cr:LiSAF has low passive losses, making it advantageous in the development of ultralow-jitter sources. However, the main drawback of these media is their low Kerr-nonlinearity.

In this thesis, we present experiments aimed at the investigation of various emerging mode locking techniques in order to generate ultrashort pulses from Cr:LiSAF and Alexandrite lasers. The first part of the thesis focuses on the use of a gain matched output coupler (GMOC) for robust and efficient Kerr-lens mode locking (KLM) of Cr:LiSAF lasers. As a pump source, a tapered diode (TD) laser with a power of 1W was utilized at 685 nm. In the experiments, it was shown that GMOC provided a significant improvement in the efficiency and robustness of the KLM operation by reducing the gain-filtering effect. The tapered diode pumped Cr:LiSAF oscillator generated pulses as short as 14.5 fs with 106 mW of average output power and 60kW of peak power at an incident pump power of 940 mW. A femtosecond tuning range extending from 807 nm to 919 nm was also achieved with sub-50-fs long pulses.

In the second part of this thesis, we present the femtosecond pulse generation experiments from a multipass-cavity Alexandrite laser. First, to the best of our knowledge, we demonstrated the generation of the shortest femtosecond pulses reported to date from a Kerr-lens mode-locked Alexandrite laser operating near 750 nm. A continuous-wave, 532-nm laser was used as a pump source and the performance of both the short and extended resonators was investigated. We increased the optical path length of the short-cavity Alexandrite resonator with a multipass-cavity extension to scale up the pulse energy and to eliminate the multi-wavelength spectral instabilities observed during continuous-wave operation. The resulting increase in the

intracavity pulse energy provided enhanced Kerr-nonlinearity and eliminated the Q-switching instabilities during mode-locked operation. The Kerr-lens mode-locked MPC Alexandrite laser produced nearly transform-limited, 70-fs pulses at a pulse repetition rate of 5.6 MHz with only 1W of pump power. The time-bandwidth product was further measured to be 0.331. We then present the graphene mode-locked operation of a femtosecond Alexandrite laser at 750 nm. We adopted a resonator configuration similar to what was previously used for Kerr-lens mode locking experiments. We used a monolayer graphene saturable absorber to obtain mode locking with a pump power of 5W at 532 nm. The resonator generated nearly transform-limited, 65-fs pulses with a time-bandwidth product of 0.319 at a pulse repetition rate of 5.56 MHz. During mode-locked operation of the cavity, the output power was around 8 mW, corresponding to a pulse energy and peak power of 1.4 nJ and 22 kW. These experiments further show that graphene can be used to initiate mode locking at wavelengths as low as 750 nm.

We believe that the results described in this thesis will pave the way for the development of low cost and efficient alternative femtosecond laser sources with a wide tunability around 800 nm, a wavelength range which is important for various scientific and technological applications.

Özet

Alexandrite ($\text{Cr}^{3+}:\text{BeAl}_2\text{O}_4$) ve Cr:LiSAF ($\text{Cr}^{3+}:\text{LiSrAlF}_6$) önemli yakın kızılaltı kazanç ortamlarıdır. Bu kazanç ortamları 10 fs ve altı darbe üretimini destekleyecek genişlikte, 700 nm'den 820 nm'ye ve 780 nm'den 1110 nm'ye olmak üzere dalgaboyu ayarlanabilirlik bandına sahiptir. Az maliyetli kırmızı diyotlarla direkt olarak uyarılabilmelerinin yanında ışıma kesiti-yaşam ömrü çarpımlarının Ti:safir ($\text{Ti}^{3+}:\text{Al}_2\text{O}_3$)'den büyük olması sebebiyle bu kazanç ortamları daha düşük eşik değerlerine sahiptirler. Yüksek kuantum verimlilikleri ve performans katsayıları nedeniyle bu kazanç ortamları düşük maliyetli, gürbüz, kompakt ve verimli laser sistemleri olarak 800 nm civarında Ti:safir'e alternatif olarak sunulabilirler. Özellikle Alexandrite, üstün termo-mekanik özellikleri nedeniyle yüksek güçlü lazer uygulamaları için uygundur. Cr:LiSAF ise düşük pasif kayıp seviyesi ile ultra-düşük gürültülü kaynakların geliştirilmesi için uygundur. Diğer yandan bu kazanç ortamlarının temel dezavantajı, düşük Kerr-doğrusalsızlığına sahip olmalarıdır.

Bu tezde, Cr:LiSAF ve Alexandrite lazerlerinden femtosaniye darbeler elde etmek için çeşitli kip kilitleme tekniklerinin incelendiği deneyler sunulmaktadır. Tezin ilk kısmında, daha gürbüz ve verimli Kerr-odaklı kip kilitleme elde etmek için kazancı eşleştirilmiş çıkış aynası (GMOC) kullanımı incelenmektedir. Pompa kayağı olarak 685 nm'de 1W güç verebilen daralan kılavuz yapıları diyot kullanılmıştır. Deneylerde, GMOC'un kazanç filtreleme etkisini minimize ederek önemli ölçüde KLM verimini ve gürbüzlüğünü iyileştirdiği gösterilmiştir. Daralan kılavuz yapıları diyot ile 940 mW'da uyarılan Cr:LiSAF lazeri ortalama gücü 106 mW olan 60kW tepe güçlerine sahip 14.5 fs uzunluğunda darbeler üretmiştir. Bu lazerin dalgaboyu, 50-fs altı darbe uzunlukları ile 807 nm'den 919 nm'ye kadar ayarlanabilmiştir.

Tezin ikinci kısmında, çoklu yansımali rezonatör yapıları Alexandrite lazerinden femtosaniye darbe üretimi sunulmaktadır. Bilgimiz dahilinde, Kerr-odaklı kip kilitli rejimde 750 nm civarında çalışan Alexandrite lazerinden bugüne kadar elde edilen en kısa darbelerin üretimini göstermiş bulunmaktayız. 532-nm çıkışlı sürekli dalga lazer kaynağı ile uyarılan sistemin kısa ve uzun rezonatör performansı incelenmiştir. Darbe enerjilerini artırmak ve sürekli dalga rejiminde gözlenen spektral düzensizlikleri ortadan kaldırmak için optik yol uzunluğu, çoklu yansımali rezonatör yapısı ile artırıldı. Bunun sonucunda ulaşılan rezonatör içi enerji seviyeleri Kerr-doğrusalsızlığını artırarak kip kilitli rejimde Q-anahtarlama düzensizliklerinin görülmesini engellemiştir. Kerr-odaklı kip kilitlemeli çoklu yansımali

rezonatör yapılı Alexandrite lazeri 5.6 MHz tekrarlama frekansında sadece 1W ile neredeyse dönüşüm limitli, zaman-bant genişliği çarpımı 0.331 olan 70-fs uzunluğunda darbeler üretmiştir. Sonraki deneysel çalışmamızda ise Alexandrite lazerinin 750 nm’de grafen ile kip kilitlemeli çalıştırılışı sunulmaktadır. Kerr-odaklı kip kilitleme çalışmamızdakine benzer bir rezonatör yapısı kullanarak grafen doyabilen soğurucusu kullanarak kip kilitleme elde etmek için 532 nm’de 5W pompa gücü kullandık. Rezonatör, 5.56 MHz tekrarlama frekansında neredeyse dönüşüm limitli, zaman-bant genişliği çarpımı 0.319 olan 65 fs uzunluğunda darbeler üretti. Rezonatörün çıkış gücü, kip kilitlemeli çalıştırılışı sırasında 8 mW civarındaydı. Bu değer 1.4 nJ darbe enerjisine ve 22 kW tepe güçlerine karşılık gelmektedir. Sunulan deneysel çalışmalar, grafenin 750 nm kadar düşük dalgalarda kip kilitleme başlatmak için kullanılabileceğini göstermiştir.

Bu tezde anlatılan deneysel çalışmalar, 800 nm civarında dalga boyu ayarlanabilir, düşük maliyetli ve verimli alternatif kaynakların geliştirilmesinin önünü açacaktır. Bu dalga boyu aralığı birçok bilimsel ve teknolojik uygulama için önem arz etmektedir.

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Nomenclature

ΔN_0 : small signal inversion	28
ΔE : activation energy.....	38
Δk : wavenumber mismatch	6
ΔN : population inversion.....	26
A_{21} : Einstein's A coefficient.....	25
A_{eff} : effective area of the mode.....	16
BP: black phosphorus	75
c : speed of light in vacuum.....	11
C_m : amplitude of the mth mode	6
CNT: carbon nanotube	67
d : effective optical length of the resonator	11
d : effective second-order nonlinear coefficient.....	6, 11
D_2 : second order cavity dispersion	24
d_g : length of the gain medium	28
DAPI: 4-6-diamidino-2-phenylindole dihydrochloride	57
E_F : Fermi energy level	71
E_i : ith component of the electric field	13
ESA: excited-state absorption	37
FOM: figure of merit.....	35
$g(v)$: lineshape function	25
g_1 : degeneracy factor of the state 1	25
g_2 : degeneracy factor of the state 2	25
GDD: group delay dispersion.....	19
GMOC: gain matched output coupler	34
GSA: graphene saturable absorber	75
h : the Planck's constant.....	25
$I(t)$: instantaneous pulse intensity	15
I : intensity of the electromagnetic wave.....	25

$I^{2\omega}$: intensity of the generated second-harmonic beam	6
I^ω : intensity of the fundamental beam.....	6
I_S : small signal saturation intensity.....	28
I_{sat} : saturation intensity of the absorber.....	15
k : Boltzmann's constant.....	38
k : wave vector	69
L : length of the gain medium.....	6
l : length of the resonator	7
l_p : prism spacing.....	21
m : number of semicircular arcs completed by circulating beam.....	32
M^2 : M-square beam parameter.....	1
MPC: multipass cavity	32
N : photo-induced carrier density	72
n_0 : background refractive index	1
N_1 : population density of the energy level 1	25
n_2 : nonlinear refractive index coefficient of the Kerr medium	14
N_2 : population density of the energy level 2.....	25
n_a : absorption probability.....	4
N_g : ground state population.....	27
n_g : group index.....	7
N_s : photo-induced carrier density.....	72
N_t : total atomic density	28
P : optical path length	21
P_{ave} : average power	4
P_i : i th component of the polarization.....	13
$q(z)$: complex parameter of a Gaussian beam.....	30
QD: quantum dot.....	75
$R(z)$: radius of curvature of a Gaussian Beam	30
r : height of the paraxial ray with respect to the optical axis	29
R : radius of curvature of the mirror.....	32
R_1 : reflection coefficient of the mirror 1	28
R_2 : reflection coefficient of the mirror 2	28
RF: radio-frequency	82
$s(t)$: modulation of the saturable absorber.....	15

s_0 : unsaturated loss.....	15
SA: saturable absorber	67
SESAM: semiconductor saturable absorber mirror.....	67
SPM: self phase modulation.....	22
T: transmission	29
T_R : cavity round-trip time	17
TD: tapered diode.....	34
TI: topological insulator.....	75
TMD: transition metal dichalcogenide	75
TOD: third order dispersion.....	19
v_p : phase velocity.....	17
$w(z)$: spot size function of a Gaussian beam	30
W: intracavity energy	24
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w_L : laser spot-size	29
w_p : pump spot size.....	29
W_p : pumping rate.....	28
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z_f : location of the beam waist.....	31
α : absorption coefficient.....	72
α : fine structure constant.....	71
α_L : cavity loss	28
α_{NS} : nonsaturable absorption	72
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β : angular spread of the beam	21
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γ : self amplitude modulation coefficient	16
γ_0 : small signal differential gain coefficient.....	28
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η_a : fraction of the pump power absorbed by the gain medium	29
η_{SHG} : second harmonic conversion efficiency	5
θ : slope of the paraxial wave with respect to the optical axis.....	29
λ : vacuum wavelength.....	1
ν_{FSR} : free spectral range.....	11

v_g : group velocity	17
σ_a : absorption cross section	25
σ_e : stimulated emission cross section	25
τ : pulse duration.....	4
τ_0 : initial pulse width of the beam.....	19
τ_f : fluorescence lifetime	27
τ_g : group delay.....	19
τ_{nr} : nonradiative lifetime	38
τ_{nr}^0 : nonradiative lifetime at low temperatures.....	38
τ_r : radiative lifetime	38
ϕ_m : phase of the mth mode	6
$\chi_{ijk}^{(2)}$: second order susceptibility tensor.....	4
χ_{ijkl} : linear dielectric susceptibility tensor	13
$\chi_{ijkl}^{(3)}$: third order susceptibility tensor	4
Ω : longitudinal mode spacing	6
ω_0 : beam waist.....	31
Ω_0 : reference frequency	6
Ω_g : gain bandwidth	17

Dedicated to Sultan Cihan, Şeref Cihan, and Canan Cihan...



Chapter 1

Introduction

The theory of stimulated emission was developed by Einstein in 1916 [1]. In 1954, Townes *et al.* demonstrated the first successful stimulated emission by employing an inverted population in ammonia at microwave frequencies [2]. They coined the name maser to represent microwave amplification by stimulated emission of radiation. In the late 1950s, many different types of masers were constructed. In the meantime, the extension of stimulated emission into the optical frequencies was being considered by many scientists. The first experimentally successful optical maser or laser (light amplification by stimulated emission of radiation) device was the flashlamp-pumped ruby laser at 694 nm, reported by Theodore H. Maiman at the Hughes Research Laboratories in 1960. By the end of 1960, many different laser types were discovered, such as helium-neon, semiconductor and Nd:YAG [3-5]. In 1964, Townes, Basov and Prokhorov received the Nobel Prize in Physics for their contributions to the development of oscillators and amplifiers based on the maser-laser principle.

The generated light waves from a laser source have some important characteristics such as, monochromaticity, coherence, brightness and directionality. For example, laser beams remain in focus (collimated) due to their spatial coherence. The distance over which a laser beam remains collimated is called the Rayleigh range and is determined for a Gaussian beam from the equation 1.1.

$$z_0 = \frac{n_0 \pi \omega_0^2}{M^2 \lambda} \quad (1.1)$$

where n_0 is the refractive index of the medium, λ is the vacuum wavelength and M^2 is the value that quantifies the deviation of the beam from its ideal diffraction-limited situation.

Pulsed lasers operating in ns regime have various applications. For example, they are utilised in spaceborn, airborne and stationary LIDAR systems for remote sensing, 3D mapping, atmospheric and terrestrial measurements. One of the first applications of the laser was optical ranging, in which the optical echoes were received from the moon by Smullin and Fiocco at MIT [6]. Based on the time between transmitted and received laser signals, the range of the target can be accurately obtained. Laser sources can be used in pulsed or continuous wave regimes in LIDAR systems. The amplitude and the phase information of the received signals reveal the valuable information about the target or the medium of interest. The parameters of the laser sources depend on the application of the LIDARs. For example the topography of the surface of the Mars was obtained by the Mars Orbiter Laser Altimeter (MOLA) which used infrared laser pulses at a repetition rate of 10 Hz [7]. In differential-absorption lidars (DIALs), generally near-infrared wavelengths are preferred due to the absorption lines of the molecules of interest. In Raman LIDARs, shorter wavelengths (320-550 nm) are used due to the wavelength dependence of the Raman scattering ($\sim \lambda^{-4}$) [8].

The invention of broadband vibronic lasers with ultrafast modulation techniques, such as Kerr lens mode-locking and saturable absorber mode-locking paved the way to the development of mode-locked lasers with pulse widths on the order of femtosecond and picosecond time scales [9-16]. Ultrafast laser technology enabled very high temporal confinement of the energy which resulted in extreme high peak powers without necessarily having high average powers. In this regard, mode-locked lasers are very crucial tools for a variety of applications in many areas of science and technology. Here, we will briefly outline some examples for applications of the ultrafast mode-locked lasers.

One important application of ultrafast lasers is multi-photon microscopy (MPM). Denk *et al.* demonstrated MPM in 1990 [17]. To obtain high enough peak powers to enable multi-photon absorption with average intensities sufficiently low (milliwatts) to avoid optical damages, the excitation of the MPM systems are often made with a mode-locked laser. The use of ultrashort laser sources dramatically improved the detection of signal photons especially when imaging deep in highly scattering environments. In MPM, two or more low-energy photons simultaneously excite the fluorescent molecule attached to specific locations in a specimen which then emits a single fluorescence photon with higher energy. The nonlinear processes illustrated in Fig.1.1 constitute the basis of this imaging technique. The probability of observing a photon at a point r within the incremental area dA and incremental time interval dt is proportional to the intensity of the beam at r and t [18],

$$p(r,t)dAdt \propto I(r,t)dAdt. \quad (1.2)$$

Thus, the multi-photon absorption and fluorescence emission rates are proportional to higher orders of power intensity. Since the intensity of the beam is highest in the vicinity of the focus, the excitation of the fluorophores are obtained exclusively in this focal volume.

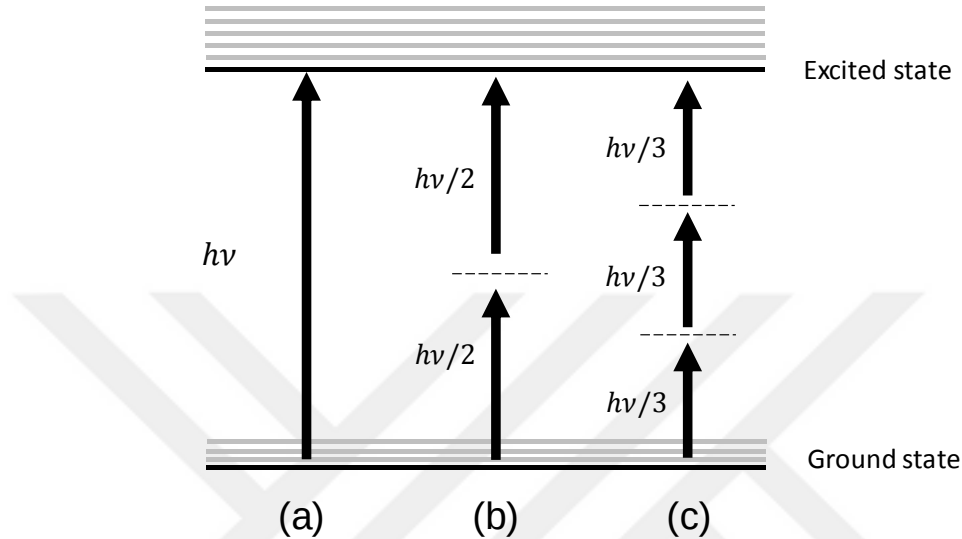


Figure 1. 1 Schematic diagram showing the absorption processes of **(a)** one-photon absorption, **(b)** two-photon absorption, and **(c)** three-photon absorption. The virtual states are represented by dashed lines.

The localization of this nonlinear excitation in the focal point leads to enhanced resolution in comparison with single-photon microscopy. The likelihood of obtaining fluorescent events occurring outside of the focal point decreases dramatically (Fig. 1.2). For this reason, there is a reduction of the background signals coming from out of focus fluorescence.

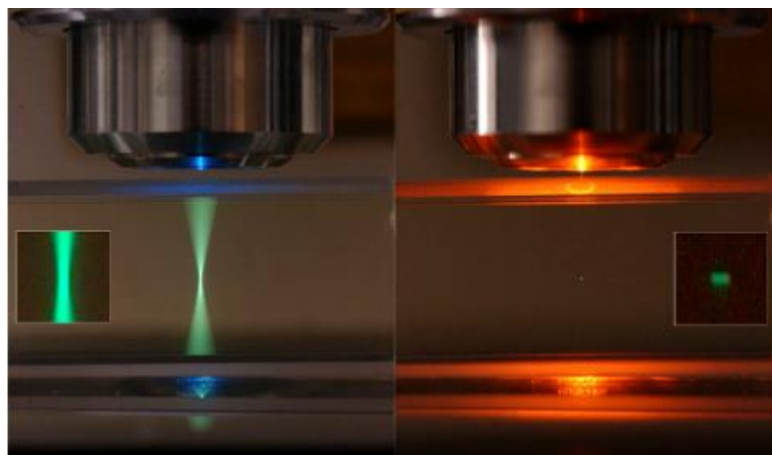


Figure 1. 2 Conventional epifluorescence (left) and two photon optical sectioning along the z-axis (right). Retrieved from [19].

Another advantage of multiphoton microscopy stems from the longer wavelength excitation which allows deeper penetration into biological tissues. For example, during the three-photon excitation (UV) microscopy, one can eliminate the disadvantages stemming from UV-matter interactions such as polymerization or temperature effects [20]. The near-infrared spectral region is a suitable window for the MPM, since, in particular, it includes the two-photon excitation wavelength range (700 nm-1000nm) of common fluorophores [21, 22].

The simultaneous absorption probability (n_a) of n photons by a certain fluorophore is proportional to several parameters of the laser pulse such as average power (P_{ave}), pulse duration (τ), repetition rate (f), and excitation wavelength (λ) [17, 20, 21, 23]

$$n_a \propto \frac{P_{ave}^n}{\tau (f \lambda)^n}. \quad (1.3)$$

According to relation given above, to enhance the multi-photon absorption, shorter pulses might be beneficial [24]. However, we encounter other challenges such as the dispersion compensation of the microscope optics for very short pulses. As the bandwidth of the pulse increases, the higher order dispersion effects become not negligible. In this case, a 10-fs pulse with a non optimum dispersion compensation could generate a weaker fluorescent signal compared to that of a 100-fs pulse [24].

The high intensity of the laser beam further leads to other optical phenomena such as high harmonic generation. Franken *et al.* reported the first generation of optical harmonics in 1961[25]. Then, Bloembergen *et al.* developed the theoretical formulation of nonlinear optics [26, 27]. Nonlinear optics deals with light-matter interactions in media where the higher-order susceptibilities $\chi_{ijk}^{(2)}$ and $\chi_{ijkl}^{(3)}$ are no longer negligible. As the strength of the applied electric field is increased, the induced polarization is not linearly proportional to the electric field, but depends on higher orders terms which can be expressed in a Taylor series expansion [28].

$$P_i = \epsilon_0 \left(\chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l + \dots \right) \quad (1.4)$$

After the invention of the lasers, nonlinear optical interactions were experimentally realized and investigated. The nonlinear optical response of the material leads to numerous interesting

phenomena and useful applications. The second-order nonlinearity $\chi_{ijk}^{(2)} E_j E_k$ is responsible for several nonlinear interactions such as second-harmonic generation (SHG), three-wave mixing, and parametric amplification and oscillation. The third-order nonlinearity $\chi_{ijkl}^{(3)} E_j E_k E_l$ is responsible for third-harmonic generation (THG), the Kerr effect, four-wave mixing, etc. [29]. For simplicity, we will only consider the example of second harmonic generation which is illustrated in Fig.1.3.

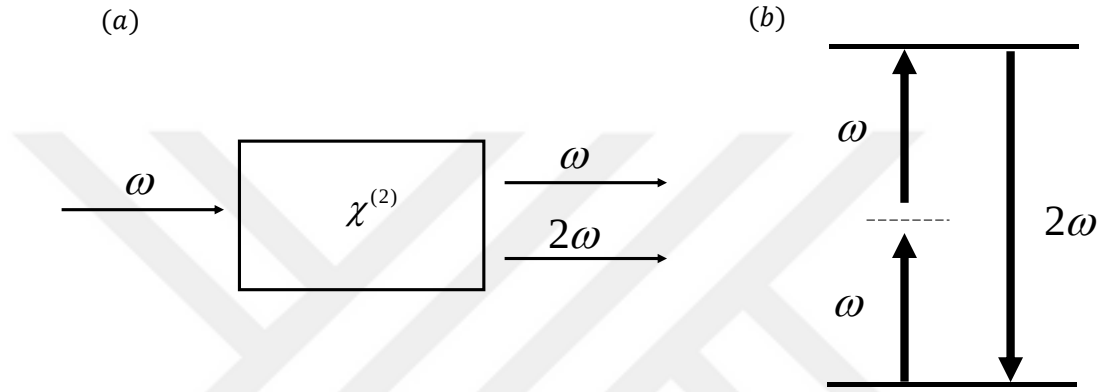


Figure 1. 3 (a) Geometry of second-harmonic generation, (b) Energy-level diagram describing second-harmonic generation [30].

The interacting two waves, with frequencies ω_1 and ω_3 ($\omega_3 = 2\omega_1$) with the second order nonlinearity can be expressed as

$$E_i^{\omega_1}(t) = \frac{1}{2} (E_{0i}^{\omega_1} e^{j(\omega_1 t - k_1 z)} + c.c.), \quad (1.5)$$

$$E_i^{\omega_3}(t) = \frac{1}{2} (E_{0i}^{\omega_3} e^{j(\omega_3 t - k_3 z)} + c.c.).$$

The detailed derivations can be found in the references [28, 29]. Under the assumption of the power lost from the incident beam (ω_1) is negligible during the conversion to $2\omega_1$, the second harmonic conversion efficiency η_{SHG} can be written as

$$\eta_{SHG} = \frac{I^{2\omega}}{I^\omega} = \frac{2\omega^2 d^2 L^2}{n^3} \left(\frac{\mu_0}{\epsilon_0} \right)^{3/2} \frac{\sin^2 [(\Delta k) L / 2]}{[(\Delta k) L / 2]^2} I^{(\omega)} \quad (1.6)$$

where d is the effective second-order nonlinear coefficient, Δk is the wavenumber mismatch, L is the length of crystal, I^ω and $I^{2\omega}$ are the intensities of the fundamental and the generated second-harmonic beam.

According to Equation 1.6, in order to maximize the conversion efficiency, the phase mismatch should be zero $\Delta k = 0$. Also, the incident wave should have a high intensity. This is accomplished by use of pulsed lasers which confine the energy in very short temporal durations. Further, the wave must be focused to the smallest possible area and must have the longest possible interaction length L . High harmonics extending to X-rays can be generated via an extreme nonlinear interaction between femtosecond pulses with high peak powers and the gas molecules. The high-intensity laser beam ionizes the gas molecules and accelerates the valence electrons away from the ion. As the field reverses and the oscillating electrons return to the ion, the process results in the generation of X-rays [31, 32].

Ultrashort optical pulse generation has improved our temporal and spectral resolutions tremendously in the exploration of many natural phenomena [2, 21, 33-35]. Mode-locking enables the generation of periodic train of pulses with broad spectral widths which cannot be produced with conventional electrical systems. When viewed in the frequency domain, the mode-locked laser spectrum consists of a series of equally spaced modes called "frequency combs" which allow precise frequency measurements [36]. The output pulse train of a mode-locked laser can be expressed as [29, 37]

$$E(t) = \sum_m C_m e^{j[(\Omega_0 + m\Omega)t + \phi_m]} \quad (1.7)$$

where the longitudinal mode spacing and the reference frequency are signified as Ω and Ω_0 , respectively. C_m and ϕ_m are amplitude and phase of the m th mode.

The oscillation frequencies of the resonator are assumed to be equally spaced and described by

$$\omega_m = \Omega_0 + m\Omega, \quad (1.8)$$

where m takes integer values and previously defined Ω is given by

$$\Omega = 2\pi \frac{c}{2n_g l}. \quad (1.9)$$

Here, l and n_g are the resonator length and the group index, respectively. Modulation at a frequency of Ω results in the coupling of the oscillating modes inside the resonator.

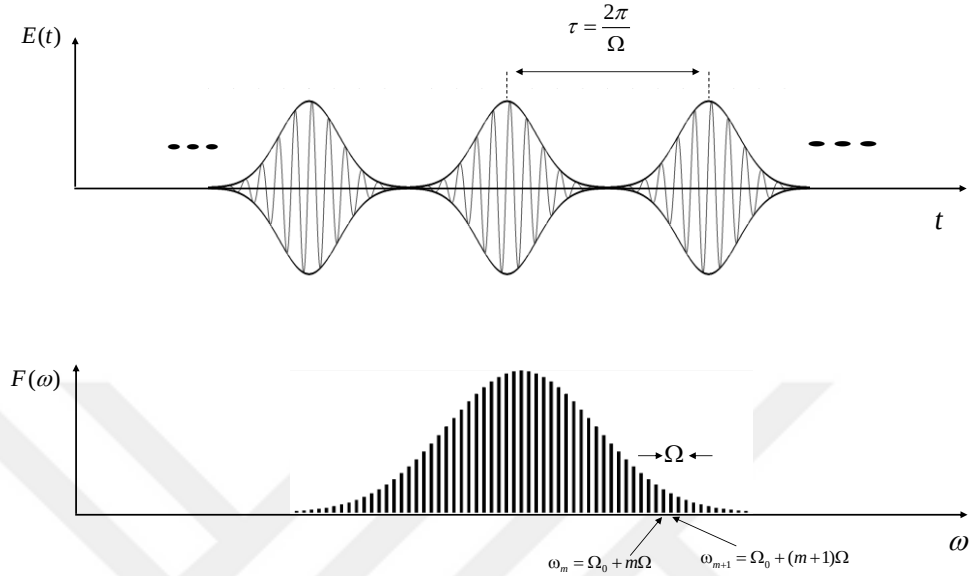


Figure 1. 4 A periodic pulse train in time and the corresponding frequency spectrum (adopted from [29, 37]).

Figure 1.4 shows the periodic pulse train and the corresponding frequency comb [29]. Mode-locked pulses in the femtosecond regime can be employed for applications in optical frequency metrology as the standard unit of time and they are expected to outperform cesium clocks. Because of their contributions to the development of laser-based precision spectroscopy, including the optical frequency comb technique, J. L. Hall and T.W. Hänsch received 2005 Nobel Prize in Physics. The applications described up to now clearly show that there is a high demand for femtosecond lasers which can produce high peak intensities and large gain bandwidths.

Tunable lasers with broad gain bandwidths are commonly used to develop femtosecond lasers. The first tunable solid state laser was reported by Johnson *et al.* from Ni^{3+} -doped MgF_2 crystals [38]. However, this laser was not practical since it required cryogenic cooling. The improvements in the tunable lasers continued with the F_A centre laser by Mollenauer and Olson in 1974 [39] and was dramatically advanced by the announcement of the Alexandrite ($\text{Cr}^{3+}:\text{BeAl}_2\text{O}_4$) laser (1979) by Walling *et al.* in Allied Corporation [40]. Alexandrite laser could operate at room temperature and was the most popular and available vibronic laser with a broad tuning range between 700-820 nm. After the invention of

Ti:sapphire laser by Moulton in 1982 [41], Ti:sapphire laser has become the most widely used tunable solid-state laser. The great interest in Ti:sapphire stems from its broadest gain bandwidth (660-1180 nm) among other solid state gain media [42]. This broad gain bandwidth allows the generation of pulses with a time duration of 5 fs [42-44]. Moreover, Ti:sapphire technology enables the production of very intense (petawatt level) pulses with the chirp pulse amplification technique [45]. Besides its broad tuning range, Ti:sapphire possesses a large gain cross section that is comparable to Nd:YAG. Further, Ti:sapphire does not suffer from excited-state absorption [46, 47]. In 1989, another vibronic laser family, Cr³⁺-doped colquiriites: Cr:LiSAF (Cr³⁺: LiSrAlF₆), Cr:LiSGaF (Cr³⁺: LiSrGaF₆), and Cr:LiCAF (Cr³⁺: LiCaAlF₆) were developed by Payne *et al.* Colquiriite family attracted a great interest since they could be directly pumped by low cost diode lasers. They opened the possibility for the design of low cost and compact femtosecond laser systems [48].

This thesis describes the development of new types of femtosecond lasers based on Alexandrite and CrLiSAF gain media. Both Alexandrite and Cr:LiSAF are Cr³⁺-doped vibronic lasers. Because of their broad tuning ranges in the near-infrared spectral region, Alexandrite and Cr:LiSAF can be utilised in the development of low-cost, robust, compact and efficient femtosecond lasers sources for numerous applications. This dissertation focuses on the ultrashort pulse generation capability of Cr:LiSAF and Alexandrite gain media which can be good alternatives to Ti:sapphire around 800 nm in certain cases. We will start Chapter 2 by reviewing ultrashort pulse generation methods. Then we briefly discuss some important concepts such as, group velocity dispersion, soliton area theorem, and population inversion conditions in a four-level atomic system. The q-preserved multipass cavity design will be summarized at the end of the chapter. Chapter 2 hence provides the important theoretical background of the experimental methods used in this thesis.

We start Chapter 3 by providing some of the important Cr:LiSAF gain medium properties. In the first part of our experimental work, we present how gain-matched output couplers (GMOCs) could be effectively utilized to obtain robust Kerr-lens mode locking operation in Cr:LiSAF laser despite its low nonlinear refractive index ($n_2 = 0.8 \times 10^{-16} \text{ cm}^2 / \text{W}$) [49]. The GMOC (0.7%) matches the gain profile of Cr:LiSAF. In that way, it provides a wavelength dependent loss inside the cavity which is not possible with regular output couplers. In the experiments, a high brightness tapered diode (1W, 685 nm) was employed as a pump source and the Cr:LiSAF laser produced sub-15 fs pulses around 850 nm with peak powers as high as 60 kW at a repetition rate of 113 MHz. The results obtained in this study demonstrate

that GMOC can be used to enhance the KLM action even in a gain medium with a low nonlinear refractive index.

The second part of the thesis is devoted to the generation of sub-100 fs pulses from a multipass-cavity (MPC) Alexandrite laser. After talking about gain medium properties of the Alexandrite, in Chapter 4, we will describe the generation of 70-fs nearly transform-limited pulses from a Kerr-lens mode locked Alexandrite laser around 750 nm [50]. In the experiments, the optical path length of the resonator was increased by using a q-preserving multipass-cavity extension. Thus, we could increase the output pulse energy and eliminate the Q-switching instabilities observed in the short cavity configuration. The MPC extension also made it possible to obtain femtosecond pulses with an available pump power of only 1W at 532 nm which in turn minimized the thermal effects. Chapter 5 starts with a brief review of optical properties of graphene as a saturable absorber. Then we present our experimental results obtained from a graphene mode-locked femtosecond Alexandrite laser [51]. By using a monolayer graphene saturable absorber, Alexandrite laser generated nearly transform-limited, 65-fs pulses at a repetition rate of 5.56 MHz with 5W of pump at 532 nm. To the best of our knowledge, this study is the first experimental demonstration of graphene mode locking of Alexandrite laser. Although in this spectral range, it becomes harder to obtain mode-locking with graphene saturable absorber due to increase of the saturation fluence, our experiments further show that graphene can be used to initiate mode locking at wavelengths as low as 750 nm. Lastly, we present our conclusions in Chapter 6.

Chapter 2

Theoretical Background

In this chapter we will outline some of the background information on several important concepts related to lasers. Chapter 2 begins with an overview of mode locking techniques. After reviewing the mode locking concept, Kerr-lens mode locking and saturable absorber mode locking will be described. We then discuss the effects of dispersion on ultrashort pulses and cover soliton area theorem. Further, the conditions for population inversion in a four-level system will be explained. We end the chapter with the design principles of a q-preserving multipass cavity which provides an effective way of obtaining high peak powers.

2.1 Methods of Femtosecond Pulse Generation

Mode-locking enables the direct generation of ultrashort pulses in picosecond and femtosecond time scales [43, 52]. It can be categorized as active mode-locking or passive mode-locking, depending on the type of the mode-locker employed. In active mode-locking, generally an externally driven amplitude modulator (AM) or phase modulator (frequency modulation (FM)) is used to obtain pulsed operation. In passive mode locking, the mode-locker exhibits some optical nonlinearities such as the nonlinear change of the refractive index in a Kerr medium or saturation of a saturable absorber. In this section, we will focus on two widely used passive mode-locking techniques, namely, Kerr-lens mode locking and saturable absorber mode locking.

After its first experimental demonstration [53], mode-locking has been utilized in various laser systems to generate ultrashort pulses [54-59]. Lasers can oscillate on many longitudinal modes with frequencies that are equally separated by the Fabry-Perot intermodal spacing. Although these modes normally oscillate independently, it is possible to couple them

and lock their phases together. In mode locking, the energy of the laser is distributed among the longitudinal modes of the resonator and relative phase of the modes are locked. Here we consider, the interference of M monochromatic waves with equal intensities and equally spaced frequencies in order to explain the phenomenon. If we assume that the spatial distribution of the mode is TEM_{00} , infinitely many longitudinal modes can be supported by the resonator at discrete frequencies given by [28]

$$\nu_q = q \frac{c}{2d} \quad (2.1)$$

where q is an integer, c and d are the speed of light in vacuum and the effective optical length of the resonator, respectively.

The spacing between two adjacent modes is called the free spectral range, ν_{FSR} , and is given by

$$\nu_{FSR} = \nu_{q+1} - \nu_q = \frac{c}{2d}. \quad (2.2)$$

The total complex field can be written in the form of a sum as in the following equation.

$$E(t) = \sum_{q=0}^{M-1} E_q \exp(j(\omega_0 + q\omega_F)t + j\phi(t)) \quad (2.3)$$

In mode locking, the modulator is operated in such a way to sustain the oscillation of the modes with a constant phase. This gives rise to the generation of periodic train of ultrashort optical pulses. When the phase factors are set to zero and the equal amplitudes are assumed for each mode, we obtain

$$E(t) = \sum_{q=0}^{M-1} E_q \exp(j(\omega_0 + q\omega_F)t) = E_0 \exp(j\omega_0 t) \frac{1 - \exp(jM\omega_F t)}{1 - \exp(j\omega_F t)},$$

$$= E_0 \exp(j\omega_0 t) \frac{\sin\left(\frac{M\omega_F t}{2}\right)}{\sin\left(\frac{\omega_F t}{2}\right)}. \quad (2.4)$$

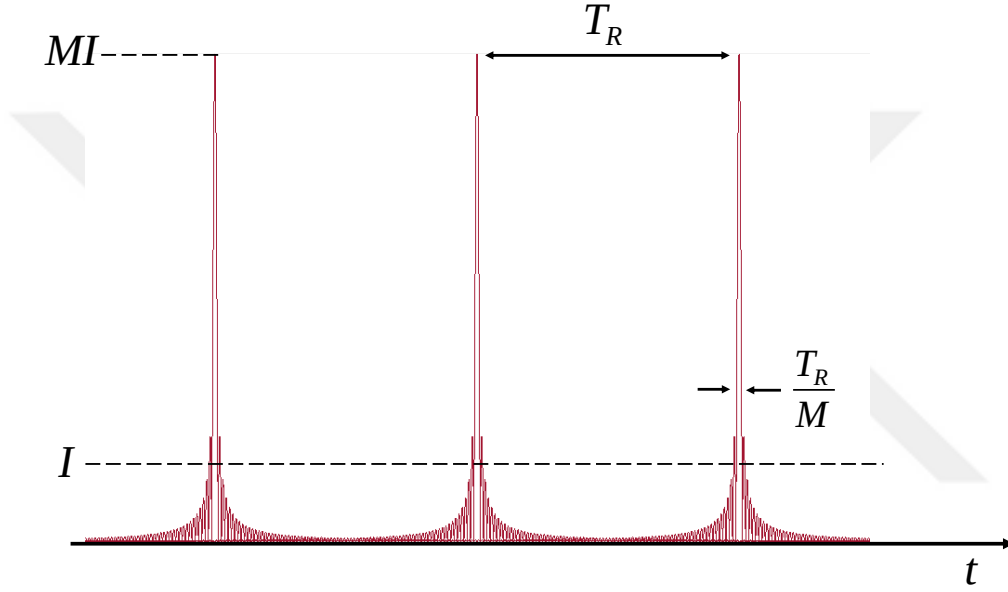


Figure 2. 1 Superposition of the M laser modes of equal amplitudes and phases resulting in a peak intensity which is M times greater than the mean intensity. Each pulse has a duration of a T_R / M .

As a result of oscillation of the modes with constant relative phase inside the cavity, the peak power of the pulses becomes M times larger than the average power. In this example the peak powers are M times larger for the coupled M modes as shown in Fig. 2.1. As the number of locked modes increases, the corresponding peak power of the mode locked laser becomes larger. If we roughly define the pulse width τ_p as the time difference between the pulse peak and the first zero, the following formulas can be used to estimate the pulse width [28]:

$$\frac{M\omega_F\tau_p}{2} = \pi,$$

$$\tau_p = \frac{1}{M} \left(\frac{2\pi}{\omega_F} \right) = \frac{T_R}{M}. \quad (2.5)$$

Another important result of mode-locking is described by Equation 2.5. As the number of locked modes increases, the pulse width decreases as depicted in Fig.2.1. After describing the mode locking concept, we will introduce two widely used passive mode locking techniques, Kerr-lens mode locking (KLM) and saturable-absorber mode locking in the following section.

2.1.1 Kerr-lens Mode Locking

One of the important results of the third-order polarization (Eqn. 2.6) is the optical Kerr effect. In response to applied field, Kerr media generates the third harmonics of frequencies. In this case the third order polarization can be described as [29]

$$P_i^{(3)} = 4\chi_{ijkl}E_jE_kE_l. \quad (2.6)$$

Here P_i and E_i are the i th component of the polarization and the electric field, respectively. χ_{ijkl} is the linear dielectric susceptibility tensor. We assume the summations over repeated indices ($i, j, k, l = x, y, z$).

The effect was first reported by J. Kerr in 1875 [60]. It is also known as the quadratic electro-optical effect. It was first utilized in the mode locking of a Ti:sapphire laser in 1991 [61]. Kerr-lens mode locking is a widely used technique to generate ultrashort pulses from various lasers [43, 62-65]. It can be considered as an artificial saturable absorber based on the nonlinear refractive index. The third-order nonlinearity results in an intensity dependent term in addition to the linear refractive index. Thus, the overall refractive index of a Kerr medium can be expressed as

$$n(r, t) = n_0 + n_2 I(r, t) \quad (2.7)$$

where n_0 is the low-intensity refractive index of the medium, n_2 is the nonlinear refractive index coefficient of the Kerr medium, and $I(r, t)$ is the intensity of the light.

The position and time dependence of the intensity creates temporal and spatial nonlinear effects inside the Kerr medium [28, 66]. Due to the spatial distribution of the optical beam, the center of the pulse at high intensity will experience a higher refractive index ($n_2 > 0$), compared to the wings of the beam. As a result, the Kerr medium will behave as a lens for $n_2 > 0$. Thus, the beam will undergo self focusing at high intensities. As the lasing volume is reduced due to self focusing, it overlaps better with the pump volume which results in higher gain experienced by the laser beam. Since the resulting high-peak pulses have higher gain in comparison with cw regime of operation, Kerr lensing induces mode-locked operation. In the temporal domain, the refractive index changes as a function of time across the temporal extend of the pulse and leads to a time varying phase. This effect is called as self-phase-modulation (SPM). The nonlinear phase shift observed after propagation in a nonlinear medium of length L is given by [67]

$$\Delta\phi(t) = -\frac{2\pi}{\lambda} n_2 I(t) L. \quad (2.8)$$

The time derivative of the total phase gives the instantaneous frequency ω_{ins} which can be expressed as

$$\omega_{ins} = \frac{d}{dx}(\omega_0 t + \Delta\phi(t)) = \omega_0 + \frac{d\Delta\phi(t)}{dx}. \quad (2.9)$$

The time-varying nonlinear phase shift obtained as a result of the SPM, results in varying frequency shifts away from the initial carrier frequency. For a medium with $n_2 > 0$, SPM gives rise to red shifts on the front edge of the pulse and blue shifts on the trailing edge. This variation of the instantaneous frequency as a function of time is called chirping. In this case, the pulse experiences an up-chirp near the center of the pulse since the frequency increases with time.

2.1.2 Saturable Absorber Mode Locking

In this part, we will overview fast saturable absorber mode locking [29, 58, 68]. Here, we consider a saturable absorber with a low saturation intensity and a relaxation time shorter than the duration of the mode locked pulses. By saturable absorber, we mean a nonlinear loss element that exhibits a low absorption for optical beams with high intensities, and a high absorption for optical beams with low intensities. The steady-state situation, occurring in this case, can be described with the help of Fig.2.2. As a result, such an absorber will favor the transmission of short optical pulses with high peak intensities in the laser resonator. We can describe the modulation $s(t)$ of the saturable absorber as follows [29, 58, 69],

$$s(t) = \frac{s_0}{1 + I(t)/I_{sat}} \quad (2.10)$$

where s_0 is unsaturated loss, $I(t)$ and I_{sat} correspond to the instantaneous pulse intensity and the saturation intensity of the absorber, respectively.

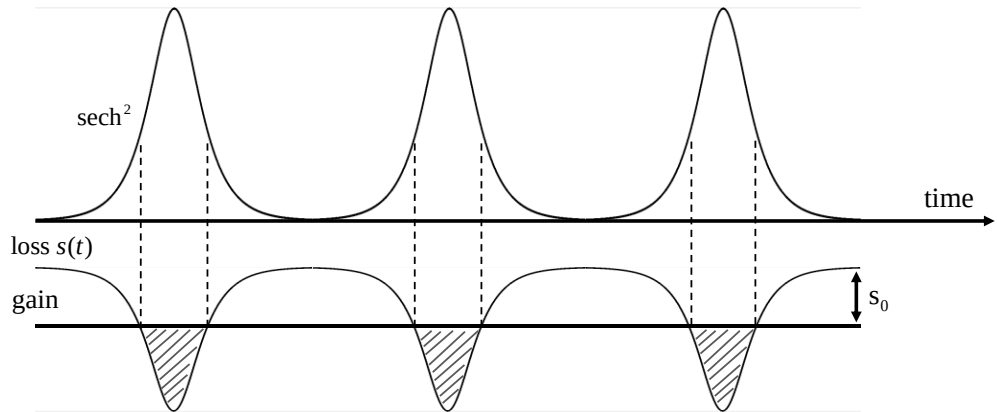


Figure 2. 2 Time variation of the pulses and the resulting saturable absorption.

If we assume weak saturation, the expression above can be expanded to give

$$s(t) = s_0 - s_0 I(t) / I_{sat}. \quad (2.11)$$

When the mode amplitude is normalized ($|E(t)|^2 = \text{Power}$), the transmission can be written as

$$\begin{aligned} T(t) &= 1 - s(t) = 1 - s_0 + s_0 \frac{|E(t)|^2}{I_{sat} A_{eff}} \\ &= 1 - s_0 + \gamma |E(t)|^2, \end{aligned} \quad (2.12)$$

where A_{eff} and γ are the effective area of the mode and the self amplitude modulation (SAM) coefficient, respectively.

Saturable absorption dynamics leads to the self amplitude modulation and the transmission which is an increasing function of the field intensity as can be seen from the Equation 2.12. As a result, the pulse evolution equation of passive mode locking with a fast saturable absorber can be written as follows [29, 58]

$$\frac{1}{T_R} \frac{\partial}{\partial T} E(t) = (g - l)E(t) + \frac{g}{\Omega_g^2} \frac{\partial^2}{\partial t^2} E(t) + \gamma |E(t)|^2 E(t). \quad (2.13)$$

The steady state solution of the equation above is a simple hyperbolic secant,

$$E(t) = E_0 \text{sech}(t / \tau) \quad (2.14)$$

where E_0 and τ are constants, with [29, 58]

$$\tau^2 = \frac{2g_0}{\gamma E_0^2 \Omega_g^2} \quad (2.15)$$

$$l - g_0 = \frac{g_0}{\Omega_g^2 \tau^2} \quad (2.16)$$

and Ω_g is the gain bandwidth and T_R is the cavity round-trip time.

In the resonator, the net gain is negative preceding and following the pulse, at low intensities, according to equation above. At the peak of the pulse, the net gain is positive due to bleaching of the saturable absorber. In this thesis, graphene was used as a fast saturable absorber. The behavior of the graphene as a saturable absorber will be further discussed in the Chapter 5 of this thesis.

2.2 Effect of Dispersion on Ultrashort Pulses and the Soliton Area Theorem

2.2.1 Phase and Group Velocities

The propagation of an electromagnetic wave can be described by two characteristic velocities, the phase and group velocities which can be used to introduce the concept of dispersion. Phase velocity v_p describes the velocity with which an observer moves to maintain a constant phase relationship with respect to a propagating sinusoidal wave and is given by

$$v_p = \frac{dz}{dt} = \frac{\omega}{k}. \quad (2.17)$$

A pulse can be regarded as a superposition of many plane waves at different frequencies. Such a superposition gives rise to a localized wave which moves at the group velocity v_g given by

$$v_g = \frac{d\omega}{dk}. \quad (2.18)$$

If the refractive index of the medium changes with the wavelength (frequency), the group velocity and the phase velocity become different of each other. In particular, the group velocity can be calculated from

$$\begin{aligned}
 v_g &= \frac{d\omega}{dk} = \frac{1}{\frac{dk}{d\omega}} = \frac{1}{\frac{d}{d\omega}\left(\frac{\omega}{v_p}\right)} = \frac{v_p}{1 - \frac{\omega}{v_p} \frac{dv_p}{d\omega}} \quad (2.19) \\
 &= \frac{1}{\frac{d}{d\omega}\left(\frac{\omega n}{c}\right)} = \frac{1}{\left[\frac{n}{c} + \frac{\omega}{c} \frac{dn}{d\omega}\right]} = \frac{c}{n + \omega \frac{dn}{d\omega}} \\
 &= \frac{c}{n - \lambda \frac{dn}{d\lambda}}.
 \end{aligned}$$

2.2.2 Effects of Group Velocity Dispersion on Ultrashort Pulses

We explained that, in a dispersive medium, different frequency components of a pulse will acquire different velocities and the pulse will broaden during the propagation. At this point, we will find the amount of the temporal spread that the pulse will experience and define the dispersion. When a pulse propagates a distance L through a medium of refraction $n(\omega)$ it accumulates the spectral phase of

$$\phi(\omega) = \frac{\omega}{c} n(\omega) L. \quad (2.20)$$

The acquired phase of the pulse can be expanded into a Taylor series around the central angular frequency [67, 70],

$$\begin{aligned}\phi(\omega) &= \sum_{j=0}^{\infty} \frac{\phi^{(j)}(\omega_0)}{j!} \cdot (\omega - \omega_0)^j \\ &= \phi(\omega_0) + \phi'(\omega_0)(\omega - \omega_0) + \frac{1}{2}\phi''(\omega_0)(\omega - \omega_0)^2 + \frac{1}{6}\phi'''(\omega_0)(\omega - \omega_0)^3 + \dots\end{aligned}\quad (2.21)$$

The first derivative $(d\phi/d\omega)$ yields the group delay τ_g and describes the delay of the peak of the envelope of the incident pulse.

$$\frac{d\phi}{d\omega} = \frac{d(kL)}{d\omega} = L \left(\frac{d\omega}{dk} \right)^{-1} = \frac{L}{v_g} = \tau_g \quad (2.22)$$

Similarly the second order $(d^2\phi/d\omega^2)$ and third order $(d^3\phi/d\omega^3)$ derivatives of the phase with respect to ω yields group delay dispersion (GDD) and the the third-order dispersion (TOD) respectively. By using the relations given in Equation 2.19, GDD and TOD can be expressed as in the equations below [28, 71].

$$\frac{d^2\phi}{d\omega^2} = \frac{L\lambda^3}{2\pi c^2} \left(\frac{d^2n}{d\lambda^2} \right) \quad (2.23)$$

$$\frac{d^3\phi}{d\omega^3} = -L \left(\frac{\lambda}{2\pi c} \right)^2 \frac{1}{c} \left(3\lambda^2 \frac{d^2n}{d\lambda^2} + \lambda^3 \frac{d^3n}{d\lambda^3} \right) \quad (2.24)$$

GDD given by the Equation 2.23, describes the frequency dependence of the group delay and leads to the temporal broadening. We can categorize the type of GDD by using the formulas given above. If $d^2k/d\omega^2 > 0$, we have normal dispersion. In this case, red tail of the pulse will go faster than the blue side. If $d^2k/d\omega^2 < 0$, we have anomalous dispersion. This time the blue components will go faster than the red components. During the propagation through a medium, the optical pulse will experience a broadening irrespective of the sign of the

dispersion. If an initially transform limited Gaussian pulse encounters a group delay dispersion, its initial pulse width (FWHM) τ_0 broadens and the output pulse width τ becomes

$$\tau = \tau_0 \sqrt{1 + \left(\ln(16) \frac{GDD}{\tau_0^2} \right)^2}. \quad (2.25)$$

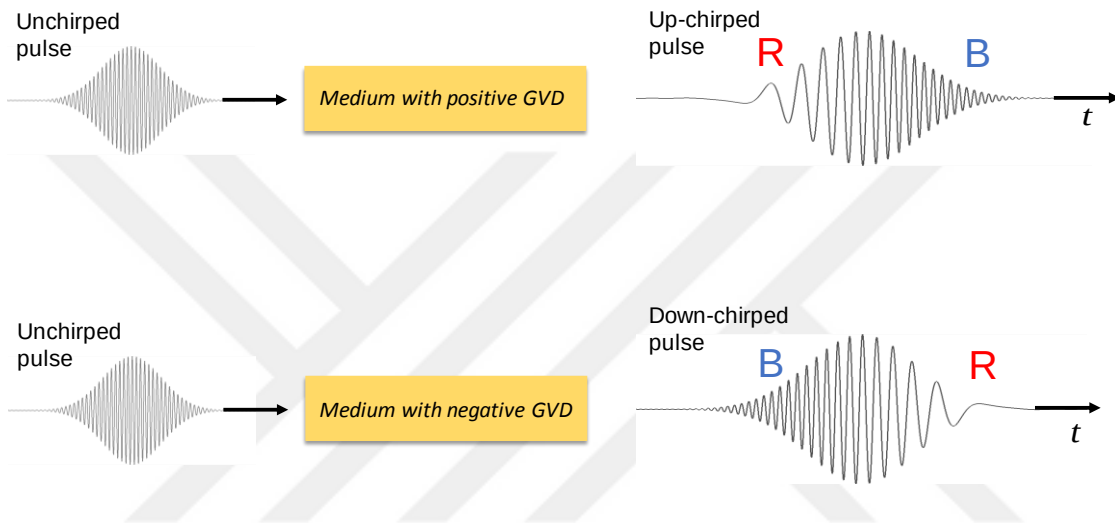


Figure 2.3 Propagation of an initially unchirped Gaussian pulse through positive and negative dispersive medium. The pulse remains Gaussian, but its width expands. It becomes up-chirped and down-chirped after positive and negative dispersive media, respectively.

Although there are other types of dispersions such as spatial dispersion and polarization dispersion [18], we will not cover all of them here. After deriving GDD, we will briefly discuss angular dispersion which arises from the frequency dependence of the refraction angle of certain optical elements such as prisms. Transparent media typically provide positive group delay dispersion in the visible region leading to up-chirped femtosecond pulses. To compress these pulses, optical systems delivering negative group delay dispersion are needed. One of the widely used optical elements for this purpose is a prism pair. Based on the angular dispersion produced by the prism pairs, the dispersion compensation to generate solitary optical pulses can be achieved. For a prism pair it can be shown that GDD is given by [67, 72, 73]

$$\frac{d^2\phi_p}{d\omega^2} = \frac{\lambda^3}{2\pi^2c^2} \frac{d^2P}{d\lambda^2} \quad (2.26)$$

$$\frac{d^2P}{d\lambda^2} = 4 \left[\frac{d^2n}{d\lambda^2} + (2n - n^{-3}) \left(\frac{dn}{d\lambda} \right)^2 \right] l_p \sin \beta - 8 \left(\frac{dn}{d\lambda} \right)^2 l_p \cos \beta \quad (2.27)$$

where P is the optical path length, l_p is the prism spacing, and β is the angular spread of the beam as shown in Figure 2.4.

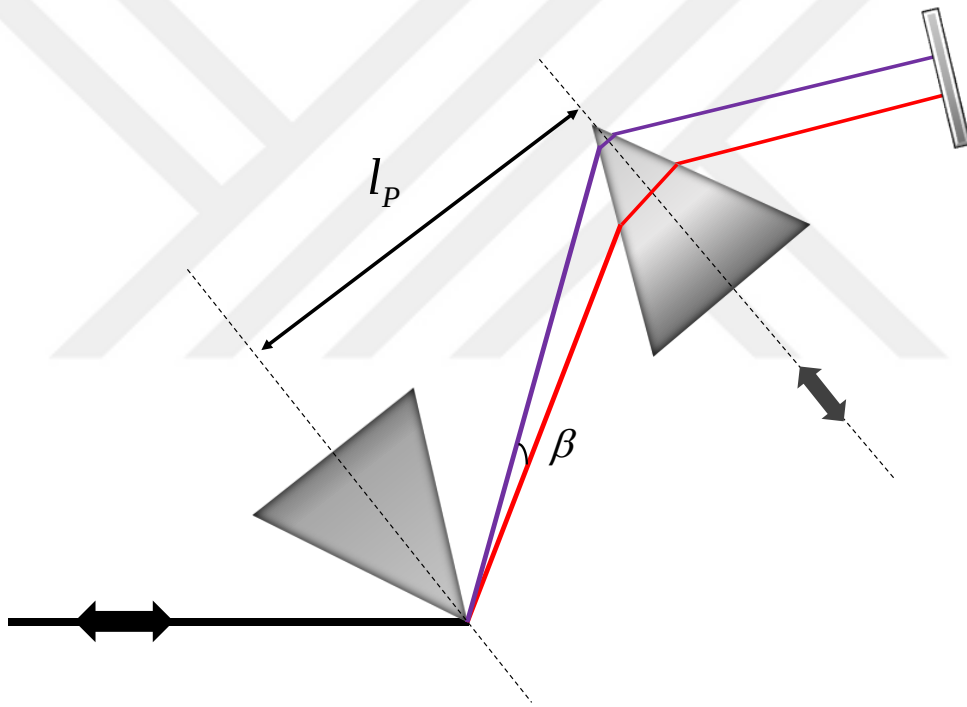


Figure 2. 4 Two-prism sequences for adjustable group delay dispersion.

In Figure 2.5, typical dispersion values per mm length of common materials, such as Cr:LiSAF, Alexandrite, and fused silica are shown. Cr:LiSAF can provide a dispersion of $\sim 24 \text{ fs}^2/\text{mm}$ around 850 nm [74] and Alexandrite can provide a dispersion of $\sim 60 \text{ fs}^2/\text{mm}$ [75] around 750 nm. Figure 2.5 also shows the GDD contribution stem from a fused silica prism pair at different separations .

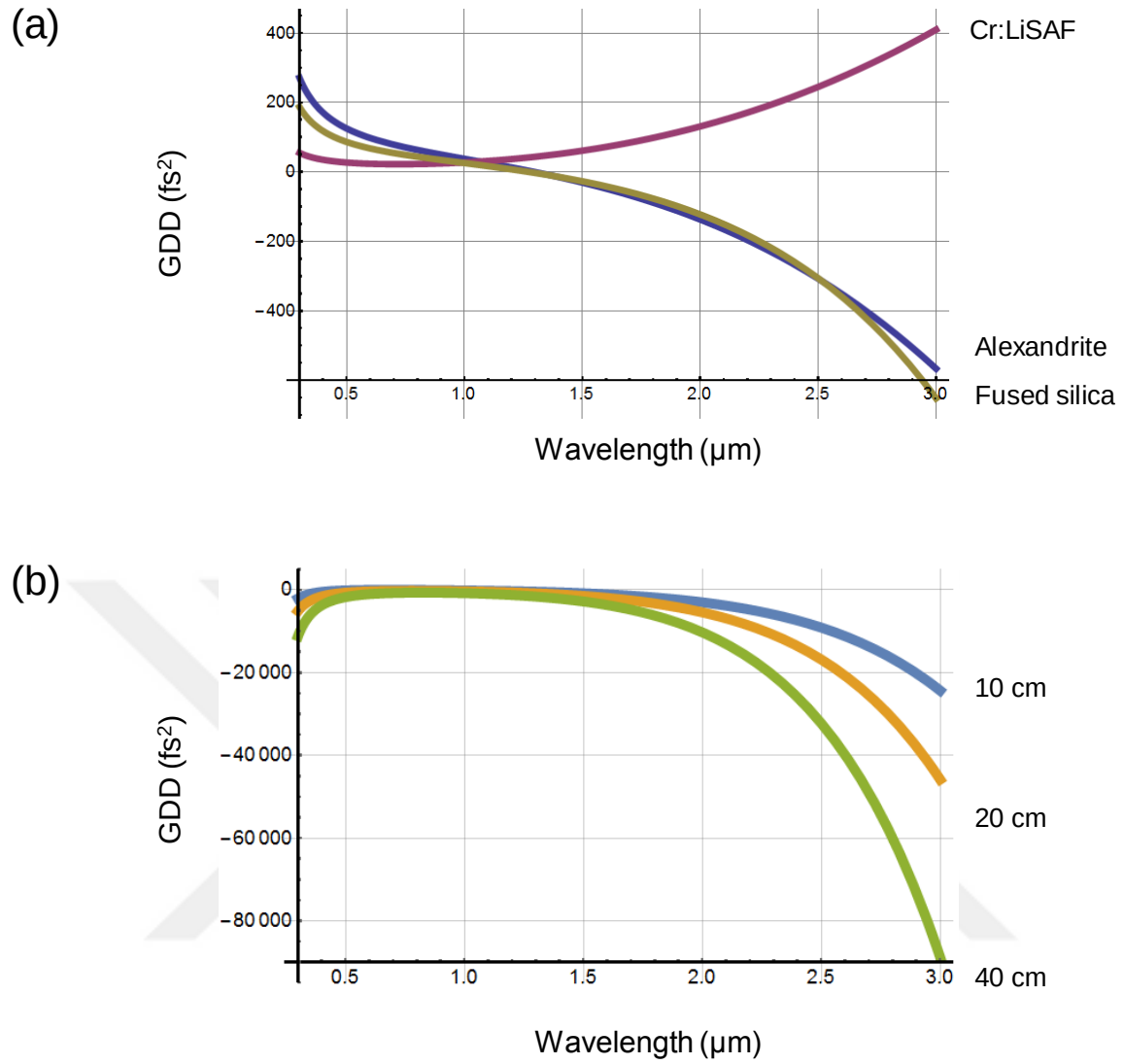


Figure 2.5 GDD introduced by different materials each of them 1mm.

2.2.3 Soliton Area Theorem

As stated previously, an optical pulse experiences self-phase modulation (SPM) due to Kerr-effect ($n = n_0 + n_2 I$) as it propagates in a nonlinear medium. The SPM leads to frequency chirping where the low-frequency components propagate faster. Under appropriate conditions, this effect can be balanced by the group delay dispersion (GDD) and solitary pulses can be

generated. When the propagating pulse experiences both SPM and GDD, the field envelope can be described by the following Nonlinear Schrödinger Equation [58, 76].

$$j \frac{\partial A(z,t)}{\partial z} = -D_2 \frac{\partial^2 A}{\partial t^2} + \gamma |A|^2 A \quad (2.28)$$

SPM and positive dispersion have similar effects on the spectral distribution of the pulse. Both of them result in pulse spreading in time. When negative dispersion acts on the pulse, the high frequency components generated by SPM have a chance to catch up with the pulse itself since negative dispersion causes high frequency components travel faster. The opposite case is valid for the low frequency components. The analytical solution of Equation 2.28 is satisfied by the following sech soliton pulse,

$$A_s(z,t) = A_0 \operatorname{sech}\left(\frac{t}{\tau}\right) e^{-j\theta} \quad (2.29)$$

where $\theta = (1/2)\gamma A_0^2 z$ gives the nonlinear phase shift of the soliton.

The phase shift must be constant over the soliton pulse with respect to time. Therefore, there must be a balance between the dispersive spreading of the pulse and the nonlinear phase shift which is written as

$$\theta = \frac{|D_2|}{\tau^2} z. \quad (2.30)$$

The integration of the squared field amplitude results in the energy of the soliton given by [58, 76]

$$w = \int_{-\infty}^{\infty} |A_s(z,t)|^2 dt = 2A_0^2 \tau. \quad (2.31)$$

Based on the above equations we obtain the soliton area theorem which relates the energy pulsewidth and the dispersion of the soliton according to

$$\tau = \frac{4|D_2|}{\gamma W}. \quad (2.32)$$

Here, W is the intracavity energy, D_2 is the second order cavity dispersion, τ is the pulse duration, and γ is the nonlinearity factor.

2.3 Population Inversion in Four-level Atomic Systems

In this thesis, four-level laser characteristics of Cr:LiSAF and Alexandrite were investigated. In the following section we will summarize the population inversion conditions and the lasing threshold conditions in a four-level atomic system.

A laser is an oscillator at optical frequencies. Light waves that become amplified through a gain medium are fed back into the resonator by reflectors and the intensity of the light beam grows. Due to the saturation effect, this intensity growth does not continue indefinitely. Before going into details of the saturation, we shall explain the optical amplification. For simplicity, consider a two level atomic system [28],

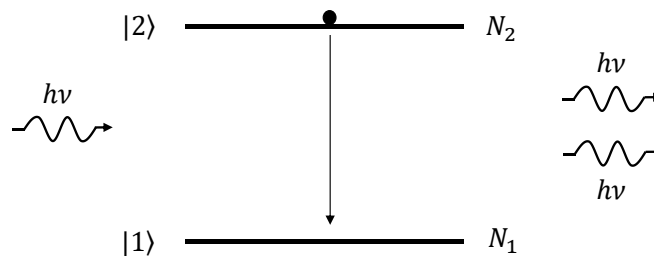


Figure 2. 6 The passage of monochromatic light beam through a two-level atomic system.

The monochromatic beam in resonance with the atomic system will cause stimulated emission, absorption, and spontaneous emission in the medium. The spontaneous emission will add noise into the emerging beam since resulting photons will be emitted in a random manner. In the following analysis, the spontaneous emission will not be considered. We can write the variation of the upper state population density as

$$\frac{dN_2}{dt} = -\sigma_e \left(\frac{I}{h\nu} \right) N_2 + \sigma_a \left(\frac{I}{h\nu} \right) N_1, \quad (2.33)$$

where N_1 and N_2 are the population densities of the energy level 1 and energy level 2, respectively. I is the intensity of the electromagnetic wave which is nearly resonant with the transition, h is the Planck's constant, σ_e and σ_a are the stimulated emission and absorption cross sections defined as

$$\sigma_e = \frac{A_{21} \lambda^2}{8\pi n^2} g(\nu),$$

$$\sigma_a = \frac{g_2}{g_1} \frac{A_{21} \lambda^2}{8\pi n^2} g(\nu),$$

where $g(\nu)$ is the lineshape function, g_1 and g_2 are the degeneracy factors of the states $|1\rangle$ and $|2\rangle$, respectively, and A_{21} is Einstein's A coefficient which gives the radiative decay rate.

The details of the analysis can be found in [28]. Here, we will review the resulting expressions for optical amplification. For a beam propagating in the z direction, the differential intensity can be expressed as:

$$\begin{aligned} \frac{dI}{dz} &= \left\{ \left[A_{21} \frac{\lambda^2}{8\pi n^2} g(\nu) \right] \left[N_2 - \frac{g_2}{g_1} N_1 \right] I \right\} \\ &= \gamma(\nu) I \end{aligned} \quad (2.34)$$

where $\gamma(\nu)$ is the differential gain coefficient given by,

$$\begin{aligned}
\gamma(\nu) &= \left(A_{21} \frac{\lambda^2}{8\pi n^2} g(\nu) \right) \left(N_2 - \frac{g_2}{g_1} N_1 \right) \\
&= \left(A_{21} \frac{\lambda^2}{8\pi n^2} g(\nu) \right) \Delta N.
\end{aligned} \tag{2.35}$$

Here, $\Delta N = (N_2 - (g_2/g_1)N_1)$ is the population inversion.

The intensity increases with distance if all terms on the right-hand side of Equation 2.35 are positive. In order to satisfy this, the population density of the upper energy level must be greater than that of the lower energy level. This can be mathematically expressed as

$$N_2 > \frac{g_2}{g_1} N_1. \tag{2.36}$$

In other words, the population inversion must be positive. In that way, the resonant light beam propagating through the atomic system will experience optical amplification. This fundamental requirement for amplification cannot be sustained under conditions of thermal equilibrium since the probability of occupancy of an energy level decreases exponentially in accordance with the Boltzmann probability distribution. Thus, we need to provide an external energy to obtain population inversion.

Now, we will analyze how the aforementioned important condition can be satisfied in a four-level atomic system. To achieve the required conditions, we will solve the rate equations in simplified four-level laser systems with steady-state laser pumping. Obtaining population inversion and low threshold laser operation is more feasible in four-level atomic systems compared to three-level and two-level atomic systems. A schematic of an ideal four-level system is shown below.

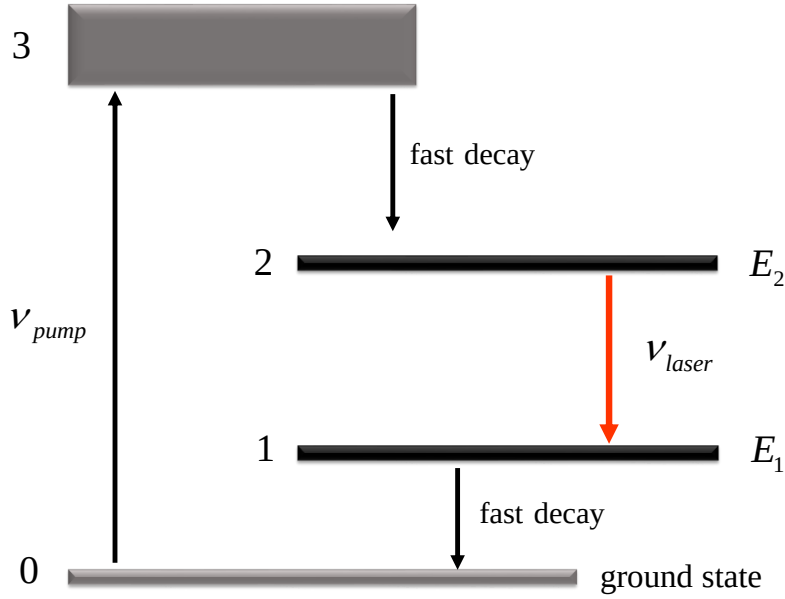


Figure 2. 7 Energy level diagram of an ideal four-level atomic system.

As shown in Fig. 2.7, a pumping mechanism excites the ground level ions into an upper level (3). Then these excited ions decay to the energy level (2) with a rapid nonradiative emission. For an ideal four-level system, the decay rates from the energy level (3) to level (2), and from the energy level (1) to the ground state (0) are much faster than the decay rate from the level (2) to the level (1). With this assumption, we can neglect the populations in levels (3) and (1) ($N_3 = N_1 \sim 0$). For this reason, the number of atoms in the energy level (2) will give the population inversion ($\Delta N = N_2$). Therefore the time rate of change of the population of the energy level (2) can be expressed as in the following rate equation,

$$\frac{dN_2}{dt} = W_p N_g - \sigma \frac{I}{h\nu} (N_2 - N_1) - \frac{N_2}{\tau_f} \quad (2.37)$$

where N_g is the ground state population, I is the intensity of the electromagnetic wave which is nearly resonant with the transition, h is the Planck's constant, τ_f is the fluorescence lifetime of the upper state, N_2 is the population density of the level 2.

The atoms are either in the ground state (0) or the level (2). Thus, we have the condition that

$$N_2 + N_g = N_t \quad (2.38)$$

where N_t is the total atomic density.

The population inversion is given by the formula below [28]

$$\Delta N = \frac{\Delta N_0}{1 + \frac{I}{I_s}} \quad (2.39)$$

where ΔN_0 and I_s are the small signal inversion and saturation intensity, respectively. They are defined as

$$\Delta N_0 = \frac{N_t W_p \tau_f}{1 + W_p \tau_f}, \quad (2.40)$$

$$I_s = \frac{h\nu}{\sigma \tau_f} (1 + W_p \tau_f). \quad (2.41)$$

The analysis provided above shows that in the presence of pumping W_p , we can always obtain the population inversion for an ideal four-level atomic system. However, to obtain net amplification, W_p should exceed a certain threshold value. The threshold condition for the light oscillation inside a resonator can be written as in the Eqn. 2.42 with the simplest resonator configuration [28]

$$(\gamma_0)_{th} = \alpha_L + \frac{1}{2d_g} \ln \left[\frac{1}{R_1 R_2} \right], \quad (2.42)$$

where R_1 and R_2 are the reflection coefficients of the mirrors, γ_0 is the small signal differential gain coefficient, and α_L represents all sources of loss inside the gain medium, d_g is the length of the gain medium.

To achieve the lasing, the laser should be pumped with a sufficient input pump power exceeding a certain threshold level. Beyond the derivation for the simplest case, the threshold pump power for our lasers in which a Gaussian beam interacts with gain medium is given by [77]

$$(P_p)_{th} = \frac{\pi h \nu_p (w_L^2 + w_P^2)(L + T)}{4 \eta_a \sigma_e \tau_f} \quad (2.43)$$

where L and T are the total resonator loss and the transmissions of the cavity mirrors, respectively; w_L is the laser spot size inside the gain medium, w_P is the pump spot size inside the gain medium, $h \nu_p$ gives the energy of the pump photons, τ_f is the fluorescence lifetime of the gain medium, σ_e is the stimulated emission cross-section, η_a corresponds to the fraction of the pump power absorbed by the gain medium.

As described by the Equation 2.43, both cavity and gain medium parameters effect the threshold pump power. Another important result is that the threshold pump power is inversely proportional to the product of stimulated emission cross-section and fluorescence lifetime ($\sigma_e \tau_f$). This is higher in Cr:LiSAF ($320 \times 10^{-20} \text{ cm}^2\text{-}\mu\text{s}$) and Alexandrite ($183 \times 10^{-20} \text{ cm}^2\text{-}\mu\text{s}$) gain media compared to Ti:sapphire ($130 \times 10^{-20} \text{ cm}^2\text{-}\mu\text{s}$). For this reason, lower threshold values can be obtained in Alexandrite and Cr:LiSAF lasers.

2.4 q-Preserving Multipass-cavity Extension

Solid state lasers that generate high energy pulses with high peak powers are widely used in various branches of science and technology [78-80]. One way of increasing pulse energies of mode locked lasers is to increase the optical path of the cavity. Multipass cavity extension is a compact and effective way of scaling up the pulse energies. The multipass cavity (MPC) was introduced by Herriot *et al* in 1964 [81]. In this part, we give a brief discussion of q-preserving multipass cavity design with the matrix formalism used in geometrical optics.

For the validity of our formalism, two important approximations need to be satisfied [82, 83]. First, we assume that the wavelength of light is negligible in comparison with the optical elements. Thus, its propagation can be described in terms of rays. Second, we consider only the paraxial rays that remain close to the axis and almost parallel to it so that the first order approximations for the sines or tangents of any angles will be involved. The methodology will not reveal the third order effects such as aberrations, astigmatism, and distortion. Two parameters of a paraxial ray (its height r and slope θ with respect to the optical axis) will be used to describe the propagation of the ray through an optical system. Therefore, this ray is described as a 2x1 column vector. An optical system will modify these ray parameters so that

the emerging ray at the output plane will have different height r_2 and different slope θ_2 and the relation of the output ray with the input ray can be written as follows [28]

$$\begin{bmatrix} r_f \\ \theta_f \end{bmatrix} = \begin{bmatrix} A & B \\ C & D \end{bmatrix} \begin{bmatrix} r_i \\ \theta_i \end{bmatrix}. \quad (2.44)$$

Here, the ABCD matrix that relates the output beam and the input beam $M_T = \begin{bmatrix} A & B \\ C & D \end{bmatrix}$ is called the ray transfer matrix. Each element in an optical system can be represented by a ray transformation matrix. In order to obtain a single matrix to represent the complete system, we multiply all the elementary transformation matrices in the correct sequence,

$$M_T = M_n M_{n-1} \dots M_1 = \prod_{i=1}^n M_i. \quad (2.45)$$

The complex parameter $q(z)$ consists of the radius of curvature $R(z)$ and the spot size function $w(z)$ of the circulating Gaussian beam and is given by

$$\frac{1}{q(z)} = \frac{1}{R(z)} - j \frac{\lambda}{n_0 \pi w^2(z)} \quad (2.46)$$

where the radius of curvature and the spot size functions are expressed as follows [28]

$$R(z) = (z - z_f) \left(1 + \left(\frac{z_0}{z - z_f} \right)^2 \right), \quad (2.47)$$

$$w(z) = w_0 \sqrt{1 + \left(\frac{z - z_f}{z_0} \right)^2}. \quad (2.48)$$

In the above equations, z is the propagation distance, w_0 is the beam waist, z_f is the location of the beam waist, and z_0 is the Rayleigh range of the beam and given by

$$z_0 = \frac{n\pi w_0^2}{\lambda}. \quad (2.49)$$

The Rayleigh range is the distance at which the initial spotsize increases by a factor of $\sqrt{2}$.

For a q-preserving resonator extension, the initial and the final q-parameters of the Gaussian beam (see Fig. 2.8) are equal at the entrance of the multipass cavity.

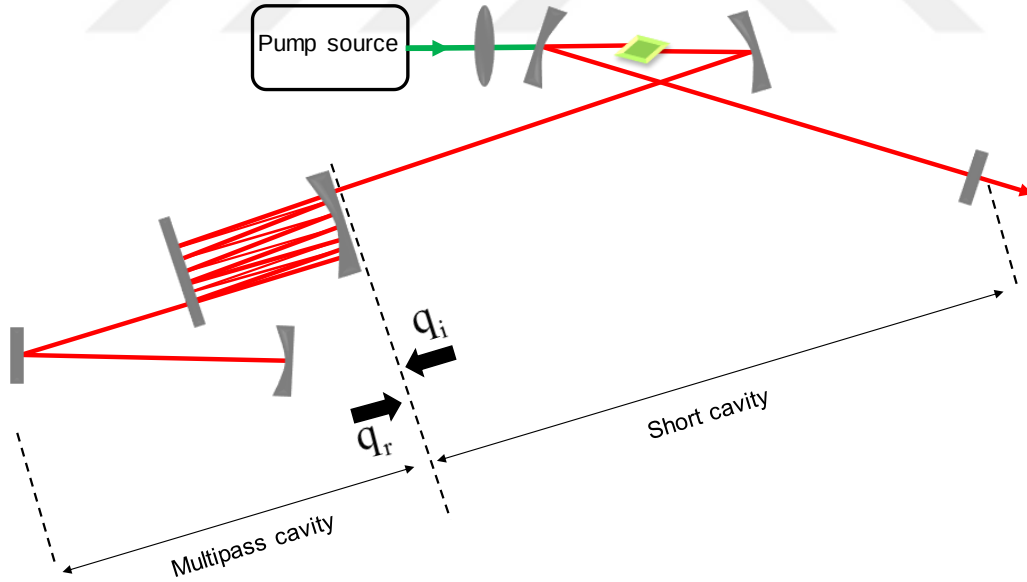


Figure 2. 8 The schematic of a multipass cavity (MPC) extended short laser cavity. The incident beam q_i and the beam returning from the q-preserving MPC q_r have the same q-parameters.

The advantage of a q-preserving multipass cavity is to provide an optical path extension without changing the initial spot size distribution of the short cavity. Thus, the intracavity energies can

be scaled up considerably while the radius of curvature and the spot size remain invariant after n number of round-trips inside the resonator extension.

Let us describe the MPC with its simplest version where we have two highly reflecting mirrors and notches for beam injection and extraction. In order to make the MPC stable, the following condition must be satisfied:

$$\left[\frac{A+D}{2} \right] \leq 1. \quad (2.50)$$

In this case, the ray transfer matrix for the MPC extension can be written in the following form [84]

$$M_T^n = \begin{pmatrix} \frac{A-D}{2} \frac{\sin n\phi}{\sin \phi} + \cos n\phi & B \frac{\sin n\phi}{\sin \phi} \\ C \frac{\sin n\phi}{\sin \phi} & \frac{D-A}{2} \frac{\sin n\phi}{\sin \phi} + \cos n\phi \end{pmatrix} \quad (2.51)$$

where, n is the number of bounces of the beam inside the MPC, ϕ is the angular advance of the spot pattern after each round-trip.

For a q preserving multipass cavity, the separation between two MPC mirrors satisfies

$$d_{MPC} = \frac{R}{2} \left(1 - \cos \left(\frac{m\pi}{n} \right) \right). \quad (2.52)$$

Here, R is the radius of curvature of the curved MPC mirror, m is the number of semicircular arcs completed by the circulating beam, and n is the number of roundtrips.

Due to the presence of the notches, the missing roundtrip also must travel a distance equal to the separation between the MPC mirrors to guarantee the q-preserving condition [85]. Figure 2.9 shows the photographs of the spot distribution on the flat (left) and curved (right) MPC mirrors of the Alexandrite laser described in Chapters 4 and 5.

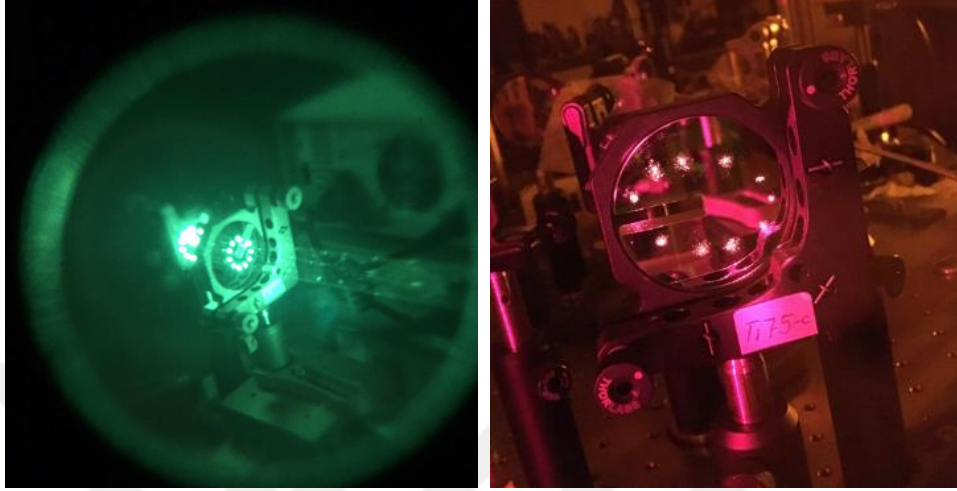


Figure 2. 9 The Photographs of the spot distribution on the flat (left) and curved (right) MPC mirrors of Alexandrite laser.

Chapter-3

Development of Femtosecond Cr:LiSAF Lasers with Gain-matched Output Couplers (GMOCs)

As stated previously, the development of low-cost, robust, compact and efficient femtosecond lasers has attracted a great deal of interest for numerous applications such as microscopy, material processing, spectroscopy, and remote sensing [86-89]. In the near-infrared spectral range, Ti:sapphire is the most widely used solid state gain medium with its broad gain bandwidth. Despite its commercial success, Ti:sapphire possesses several drawbacks. One of them is the high intrinsic losses of the Ti:sapphire crystals, on the order of 2% per cm, which results in a low figure of merit (FOM) value of 150 [90]. High intracavity passive losses also have a negative effect on the noise properties of the gain medium [91]. Moreover, Ti:sapphire lasers operate with relatively higher lasing thresholds since it has a lower product of fluorescence lifetime and emission cross section ($130 \times 10^{-20} \text{ cm}^2\mu\text{s}$) [92, 93]. Furthermore, Ti:sapphire laser systems require mainly expensive and inefficient green pump sources based on the second-harmonic of Nd and Yb-based solid state systems. This drawback has been partially eliminated, by the development of high-power laser diodes in the blue and green spectral regions [94-96]. The initial results are quite encouraging, but the demonstrated output power levels as well as the achieved optical-to optical conversion efficiency of the system require further progress. Ongoing progress in alternative pump sources for Ti:sapphire lasers are promising and may lead to the development of lower-cost and higher efficiency Ti:sapphire lasers in the future.

Cr:LiSAF has been an attractive alternative to Ti:sapphire as a broadly tunable ultrashort pulse laser source. In the following sections we will review some of the physical properties of Cr:LiSAF. Then we investigate the Kerr-lens mode-locking in a tapered-diode (TD) pumped Cr:LiSAF laser employing gain-matched output coupler (GMOC). We will present our

experimental data that demonstrates the effectiveness of the GMOC in both efficiency and robustness of KLM operation of the Cr:LiSAF laser.

3.1 Cr:LiSAF Gain Medium Properties

In the late 1980s, lasing action from a family of Cr^{3+} -doped fluorides was reported by Payne *et al.* at University of California [48]. As one of the most promising members of the Cr^{3+} -doped colquiriite family, $\text{Cr}^{3+}:\text{LiSrAlF}_6$ (Cr:LiSAF) has several attractive properties for generation and amplification of the femtosecond pulses. Cr:LiSAF possesses a broad tuning range extending from 780 to 1110 nm (see Fig. 3.1) [97]. This broad bandwidth is capable of amplifying pulses of duration ~ 10 fs or less [98, 99]. Cr:LiSAF lasers can operate with threshold values as low as 650 μW [100], due to their higher product of fluorescence lifetime and emission cross section ($320 \times 10^{-20} \text{ cm}^2\mu\text{s}$, more than 2 fold higher compared to Ti:sapphire) [45, 48]. Further, they can be directly pumped by inexpensive laser diodes around 650 nm which significantly reduces system complexity [101-104]. Moreover, Cr:LiSAF crystals can have quite low passive losses which brings an advantage in the design of low-noise femtosecond oscillators [105]. The figure of merit (FOM), the ratio of the absorption coefficients at the pump and lasing wavelengths, can be an order of magnitude better than Ti:sapphire crystals [90, 97, 106, 107]. High intrinsic slope efficiency of Cr:LiSAF (54%, [48]), allows efficient laser operation. Also, longer upper-state lifetime of 67 μs enables efficient flashlamp pumping.

On the other hand, compared to Ti:sapphire, Cr:LiSAF has a low emission cross section ($4.8 \times 10^{-20} \text{ cm}^2$) [108] and low nonlinear refractive index ($0.8 \times 10^{-20} \text{ m}^2/\text{W}$) [109]. The lower emission cross section results in lower gain. This have some consequences such as the requirement of low-loss optics and the tendency towards Q-switching. In low nonlinear refractive index gain media, Kerr-lens mode-locking becomes harder to obtain.

Table 3. 1 Comparison of the spectroscopic and laser parameters of Cr:LiSAF and Ti:sapphire.

Gain medium	Cr:LiSAF ($\text{Cr}^{3+}:\text{LiSrAlF}_6$)	Ti:sapphire ($\text{Ti}^{3+}:\text{Al}_2\text{O}_3$)
Tuning Range [nm]	780-1110 [48]	660-1180 [110]
Upper-state lifetime [μs]	67 [45]	3.2 [45]
Thermal conductivity [W/K-m]	3.09 [111]	28 [112]
$\sigma_{\text{em}}[\times 10^{-20} \text{ cm}^2]$	4.8 [109]	41 [45]
Refractive index	1.41[111]	1.76 [111]
Nonlinear refractive index coefficient [m^2/W]	0.8×10^{-20} [109]	3.2×10^{-20} [109]
$\sigma_{\text{em}}\tau [\times 10^{-20} \text{ cm}^2\text{-}\mu\text{s}]$	320 [45]	131[45]
Crystal figure of merit (FOM)	2000 [113]	150 [90]
Intrinsic slope efficiency	65 [48]	64 [114]

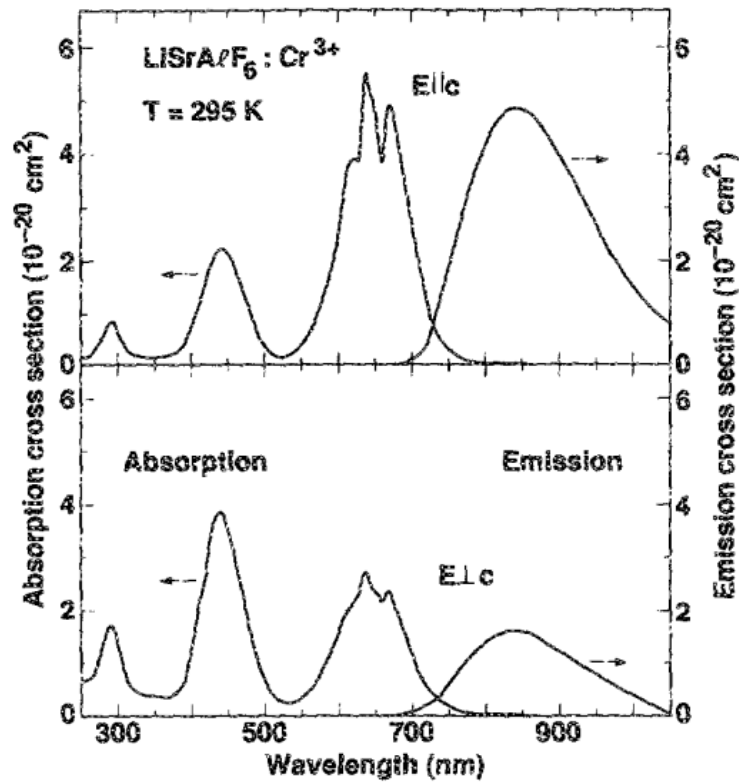


Figure 3. 1 Emission and absorption spectra of Cr:LiSAF [45, 48].

Cr³⁺-doped colquiriites also exhibit significant thermal effects that lead to a decrease in laser efficiency. If the energy from a radiative transition (photon) is converted into a nonradiative transition (phonon), it results in heating inside the crystal. There are mainly four mechanisms that give rise to nonradiative transitions in the colquiriites; quantum defect, excited-state absorption, Auger upconversion, thermal quenching (see Fig. 3.2). These undesirable effects result in decrease of the available output power [115-131].

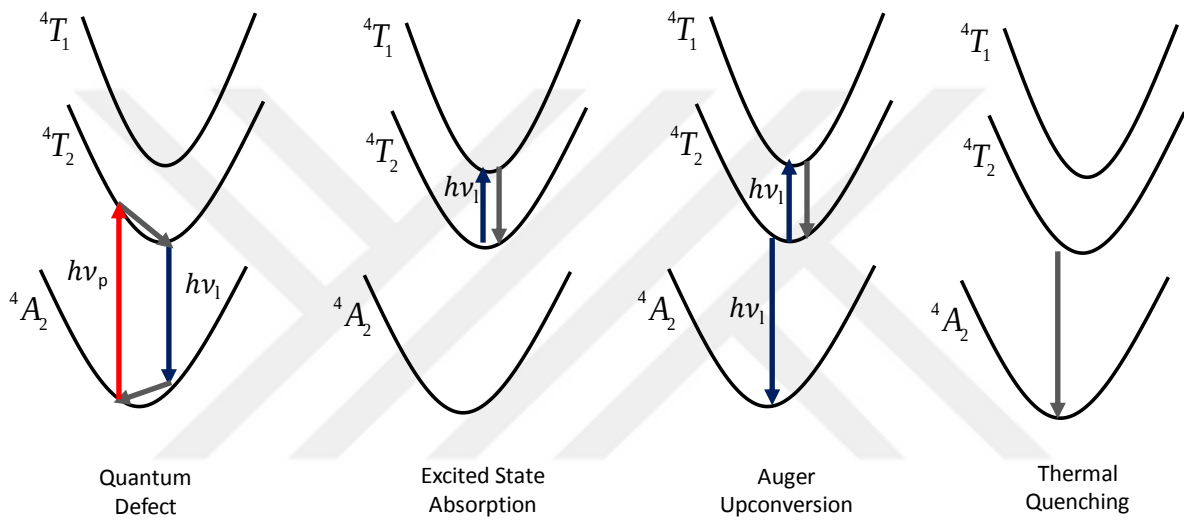


Figure 3. 2 Description of different effects that cause thermal loading in Cr³⁺-doped colquiriites.

In the quantum defect, the energy difference between the pump and laser photons ($h\nu_p - h\nu_l$) is converted into heat via nonradiative transitions inside the crystal lattice. As the pump power increases, this effect becomes predominant. However, in Cr:colquiriites, this mechanism is not a main reason for thermal loading since the difference of pump (~ 650 nm) and laser (~ 800 - 850 nm) wavelengths is small [131].

Another mechanism that leads to nonradiative transition is excited state absorption (ESA) where an ion in the upper laser level ${}^4T_2$ is excited to a higher energy level ${}^4T_1$ by a pump photon. The nonradiative decay of the excited ion back to ${}^4T_2$ level results in heating of the crystal. Furthermore, ESA does not provide a significant contribution in the thermal loading of colquiriite family due to their relatively small ESA cross sections at the lasing wavelengths [131, 132].

The Auger upconversion results from the interaction of the ions at the upper laser level $^4T_2$ where the energy created from the decay of one ion to the ground level $^4A_2$, is used to excite another ion into upper level $^4T_1$. The excited ion then nonradiatively decays back to the energy level $^4T_2$ and heats up the crystal. In the Auger upconversion, the presence of an external photons is not required. As the upper state population density increases, this mechanism could generate a significant heat load.

The final mechanism that results in thermal loading in Cr^{3+} :colquiriites is thermal quenching. The fluorescence lifetime of Cr^{3+} ions in the colquiriite host displays a significant temperature dependence [118]. At low temperatures, the upper state lifetime is nearly constant. However, as the temperature of the crystal exceeds a critical temperature, the lifetime drops rapidly. The critical temperature here is defined as the temperature at which the fluorescence lifetime drops to half of the radiative lifetime value at room temperatures. The temperature dependence of the upper state lifetime of a fluorescent ion $\tau_f(r, z)$ in a crystal is described as

$$\frac{1}{\tau_f(r, z)} = \frac{1}{\tau_r} + \frac{1}{\tau_{nr}(r, z)} \quad (3.1)$$

where

$$\tau_{nr}(r, z) = \tau_{nr}^0 \exp\left(\frac{\Delta E}{kT(r, z)}\right).$$

Here, τ_r and τ_{nr} are the spontaneous radiative and nonradiative lifetimes, respectively τ_{nr}^0 is the nonradiative lifetime at low temperatures (77K), k is the Boltzmann's constant, and ΔE is the activation energy.

The thermal quenching is the major source for heat loading among the four mechanisms described in Figure 3.2. Because, as the additional heating is generated, the rate of thermal quenching increases in an exponential manner. As a result, the performance of the laser drops dramatically. In order to eliminate the thermal quenching, the crystal temperature must be kept below the critical temperature of the gain medium. For Cr^{3+} :LiSAF, the critical temperature for thermal quenching is 69 °C.

3.2 Gain-matched Output Couplers for efficient Kerr-lens Mode Locking

Semiconductor saturable absorber mirrors (SESAMs) [133] or Kerr-lens mode-locking were utilized to generate femtosecond pulses from Cr:LiSAF laser systems [61, 134]. Although optical-to-optical conversion efficiencies of 30% could be achieved, due to their narrow operation bandwidths, SESAMs provided limited obtainable pulsewidths and tuning ranges around 25-fs and 50 nm, respectively [103, 135-138]. KLM mode-locked Cr:LiSAF systems could generate pulses as short as ~ 10 -fs [99, 139] with femtosecond tuning ranges of 835–910 and 809–910 nm [140, 141]. In Kerr-lensing, the achievable pulse durations are only limited by laser gain bandwidth and the overall dispersion of the cavity. For this reason, KLM allows generation of the shortest pulses and broadest tuning ranges among other mode-locking techniques. However, the long-term stability and robustness of the mode-locking are main concerns with KLM mode-locked Cr:LiSAF lasers because of the relatively low nonlinear refractive index (n_2) of Cr:LiSAF ($0.8 \times 10^{-16} \text{ m}^2/\text{W}$ [142]). Furthermore, the need for critical alignment of the cavity may result in lower optical-to-optical conversion efficiency in KLM Cr:LiSAF lasers depending on the pulse durations [99, 139, 143-145]. All the available gain bandwidth of the laser active medium cannot be utilised because of strong gain filtering effects which stem from wavelength dependent gain profile of the medium. This prevents generation of the shortest obtainable pulses. To overcome gain filtering effect, there is a need for large Kerr nonlinearity induced self-amplitude modulation (SAM) which requires critical cavity alignment near the edge of the laser stability region and the use of low output coupling to increase the intracavity nonlinearities. It may also result in low lasing efficiency, high mode-locking thresholds, long-term stability problems, and reduced output beam quality in ultrashort KLM lasers [145].

As the gain filtering effect is minimized, it becomes easier to maintain ultrashort pulses with KLM action and all the above mentioned problems can be further avoided. Chen *et al.* used gain matched output couplers (GMOCs) to minimize strong gain filtering effects [145]. GMOC has a transmission profile that matches the gain spectrum of the laser medium. A GMOC provides high output coupling at the high gain parts of the emission spectrum and low output coupling at the edges of the emission spectrum where the available gain is low. Hence,

a flat net gain profile is obtained over a broad spectral range. As a result, strong SAM requirement is eliminated and low level of KLM action becomes sufficient to sustain ultrashort pulses. GMOC technology was first used in a KLM Ti:sapphire system and 8-fs pulses could be generated with reduced Kerr nonlinearity requirement [145]. Later, the GMOC mirror was utilised in a low-cost, single-mode diode pumped Cr:LiSAF laser where 13-fs pulses with 25 mW of average power at an optical-to-optical conversion efficiency of 21% could be obtained [146]. The use of the GMOC provided an order of magnitude higher efficiencies compared to previous KLM Cr:LiSAF studies [99, 139, 143, 145].

In this section of the thesis, we provide our experimental data that further show the effectiveness of GMOC in obtaining efficient, stable, and robust operation of KLM Cr:LiSAF laser. The gain-matched output coupler had a transmission of around 0.7% at 850 nm and the spectral transmission profile of the GMOC matched the gain profile of Cr:LiSAF quite well on the long wavelength side as shown in Figure 3.3(a). In order to show the effectiveness of the GMOC mirror in KLM operation compared to cavities employing regular output couplers, we provide the previously obtained experimental data from a single-mode diode pumped Cr:LiSAF laser. In Figure 3.3(b), the wavelength dependence of the lasing threshold variation is shown for both the GMOC and the regular output coupler cases. To vary the wavelength of the laser, a birefringent tuning plate was used inside the cavity. Notice from Figure 3.3(b) that the GMOC provides an almost wavelength independent lasing threshold in a broad wavelength range (825–910 nm). However this is not the case with a regular OC. Threshold pump power is more wavelength dependent and steadily increasing after 850 nm with a regular output coupler due to the decrease in the gain (see Fig. 3.3(a)).

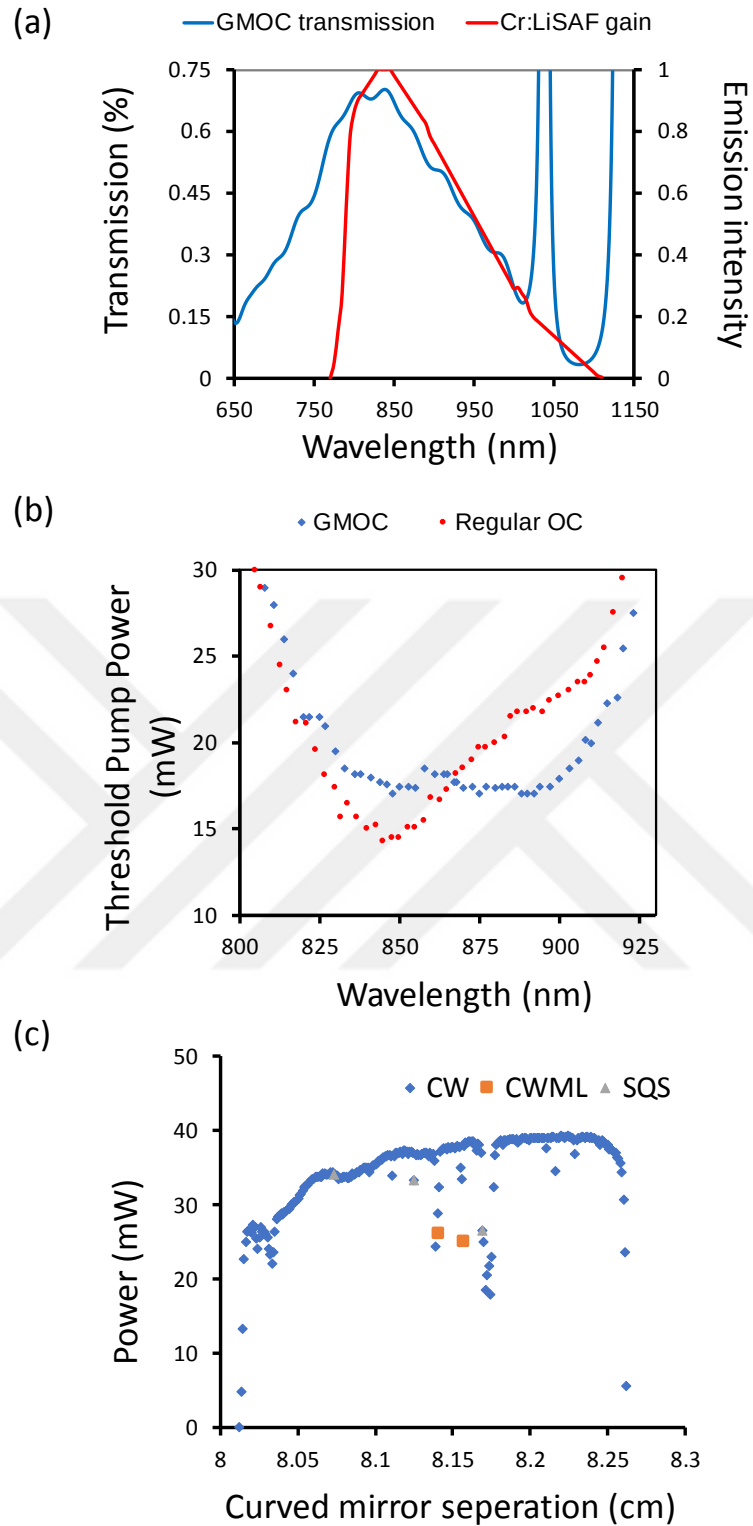


Figure 3. 3 The data taken from the single-mode diode pumped Cr:LiSAF laser system. **(a)** Designed transmission spectrum of the GMOC mirror, shown together with the wavelength dependent gain profile of Cr:LiSAF. **(b)** Measured variation of the threshold pump power required to attain lasing (P_{th}) as a function of the Cr:LiSAF laser wavelength with GMOC and regular OC . **(c)** Measured variation of the Cr:LiSAF laser output power as a function of the curved mirror separation.

The experimental results indicate that the GMOC introduces a nearly flat gain profile into the resonator by eliminating the decrease in gain due to the decreasing emission cross section. The threshold increase above the wavelengths 925 nm stems from the decreasing reflectivities of the cavity high reflectors. We expect a further extended flat net gain profile from Cr:LiSAF in the 800–1100 nm region with more optimum broadband cavity mirrors and GMOCs.

Further, the KLM operation points of the Cr:LiSAF laser was investigated across the laser stability range. During the measurements, we varied only the curved mirror separation. The Cr:LiSAF crystal position and pump focusing were not changed. Figure 3.3(c) shows the output power variation of the Cr:LiSAF laser with respect to curved mirror separation. Note that, two different operation points of stable continuous-wave mode-locking (CWML in Fig. 3.3(c)) could be initiated around the center of the laser stability range. The requirement to work near the edge of the stability region to obtain stable KLM operation was eliminated. Although the different crystal position and/or different pump focusing may provide different KLM positions, the important point to note down here is the sufficiency of even a weak Kerr-lens effect to obtain stable KLM operation with the help of the GMOC which allowed relatively good mode matching between the pump and laser modes even during KLM operation. Thus, we could obtain efficient KLM operation with high average powers. Furthermore, note that the GMOC suppresses the spectral wings of the pulses as they exit the cavity due to its output coupling profile [145]. This spectral filtering effect of the GMOC leads the output pulse durations become longer by 5–10% compared to intracavity pulse duration. For this reason, careful cavity design might be required to utilise the full spectral bandwidth of the gain medium. Self-Q-switching operation (SQS) was also observed (blue triangles in Fig. 3.3(c)), at several distinct values of the curved mirror separation. At these positions, the Cr:LiSAF laser produced pulses with a duration of several microseconds at repetition rates of several tens of kHz.

3.3 Experimental Details

Figure 3.4 shows the schematic and photograph of the Cr:LiSAF resonator used in the experiments. We used monolithic high brightness tapered diode laser (TD) to pump Cr:LiSAF system. The TD used in this work was developed at the facilities of Ferdinand Braun Institute [147]. Detailed information on this class of diodes can be found in [147]. The tapered diode provided up to 1050 mW of output power, with a diode voltage of 2.5 V and a drive current of 2 A. As a result, electrical-to-optical conversion efficiency of about 21% could be achieved. The diode spectrum was centered at 679.5 nm with a FWHM of 1.5 nm. The M^2 values of the TD was measured to be 5 and 1.1 in the slow and fast axes, respectively. The TDL had a total cavity length of 2 mm, and consisted of a 500 μm long ridge waveguide section and a 1.5 mm long tapered amplifier section with a flared angle of 3° . The output aperture was 80 μm wide. The front facet of the TDL had a reflectivity of 1%, whereas the rear facet had a reflectivity of 94%. The device was mounted p-side down on copper tungsten submounts using AuSn solder, which was then mounted on standard C-mounts. A commercial c-mount diode fixture with thermoelectric cooling was used for housing the tapered diode. The emitted power from the TD (~96%) was mainly originating from the central lobe of the beam waist. The measured astigmatism of the diode was found to be 650 μm . The parameters of the diode mentioned above provided a brightness of $B = 450\text{mW}/\mu\text{m}^2$ which was around two times higher than the previously used single-mode diodes.

Initially, an astigmatically compensated folded x-cavity was built by using two curved pump mirrors (M1 and M2, $R = 75\text{mm}$), with a reflectivity above 99.9% from 790 to 940 nm and a transmission above 98% from 625 to 685 nm to transmit the pump beams. The output beam of the diode was focused into a 6-mm-long, Brewster-cut, 1.5% Cr doped Cr:LiSAF crystal by using an achromatic doublet ($f_2=60\text{mm}$). The collimation of the diode was varied inside the Cr:LiSAF crystal with a collimating lens ($f_1=4.5\text{ mm}$) to match the laser cavity mode. Furthermore, the divergence of the beam along the fast and slow axes was controlled by a cylindrical lens ($f_z=50\text{ mm}$). The absorption of the crystal was measured to be 93% at the pump wavelength of 680 nm. During the experiments, the gain medium was surrounded by an indium foil and kept in a water-cooled copper holder at 15°C .

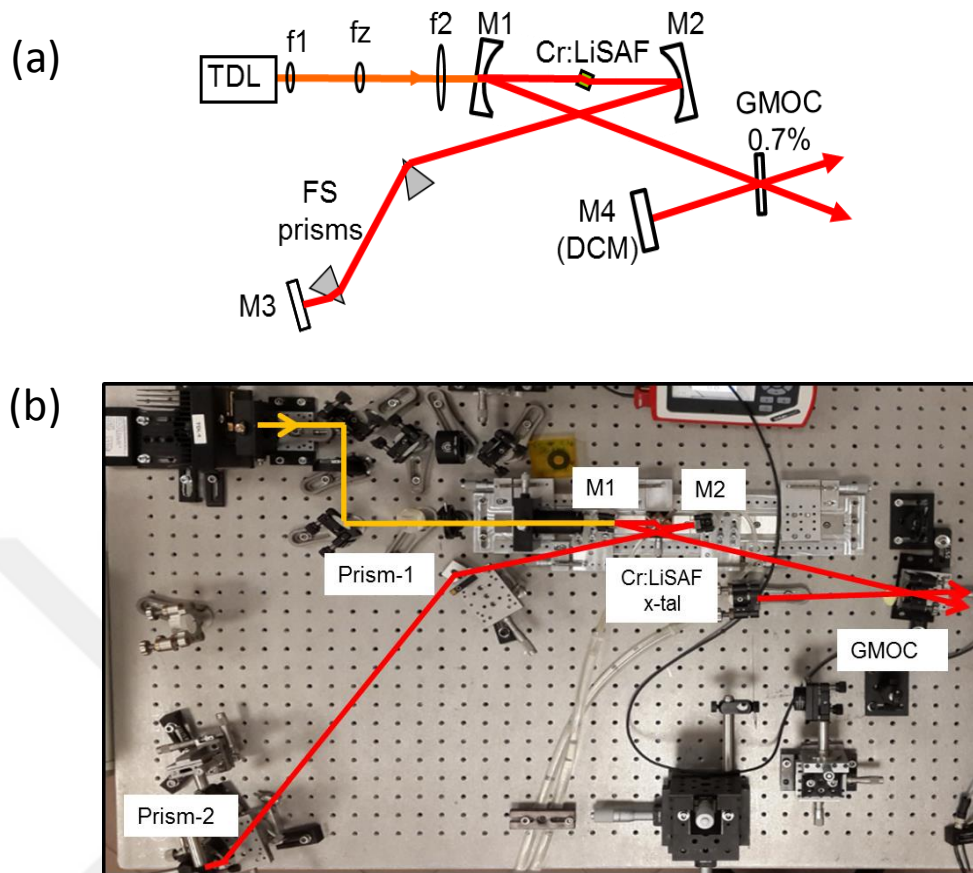


Figure 3. 4 (a) Schematic and (b) photograph of the tapered diode (TD) pumped Kerr-lens mode-locked Cr:LiSAF lasers. GMOC: Gain-matched output coupler. FS: Fused-silica.

The obtainable output pulse energies were limited by intracavity nonlinearities stem from the low transmission of the GMOC mirror. In order to obtain higher pulse energies from the TD pumped system, we increased the total output coupling to 1.4% by using a double bounce configuration on the GMOC mirror as shown in the Figure 3.4. Double-chirped mirrors (DCMs) and fused silica prism were used for dispersion compensation. The fused silica prism pairs were separated by 45 cm.

3.4 Experimental Results

In this section, experimental results obtained from the TD pumped KLM Cr:LiSAF laser are presented. We could investigate the power scalability of femtosecond Cr:LiSAF laser system due to higher brightness and higher average powers provided by the tapered diode. Figure 3.5 shows the measured power efficiency data of the Cr:LiSAF laser in CW and CWML regimes taken with the GMOC. Near the center of the outer stability range, we optimized continuous-wave (cw) laser power and obtained the CW (Optimum) data. For this case, the measured cw lasing threshold (160 mW) and laser slope efficiency (34 %) of the Cr:LiSAF laser are presented in Fig. 3.5. For an absorbed pump power of 875 mW, the laser provided a cw output power of 232 mW (116 mW from each) around 850 nm.

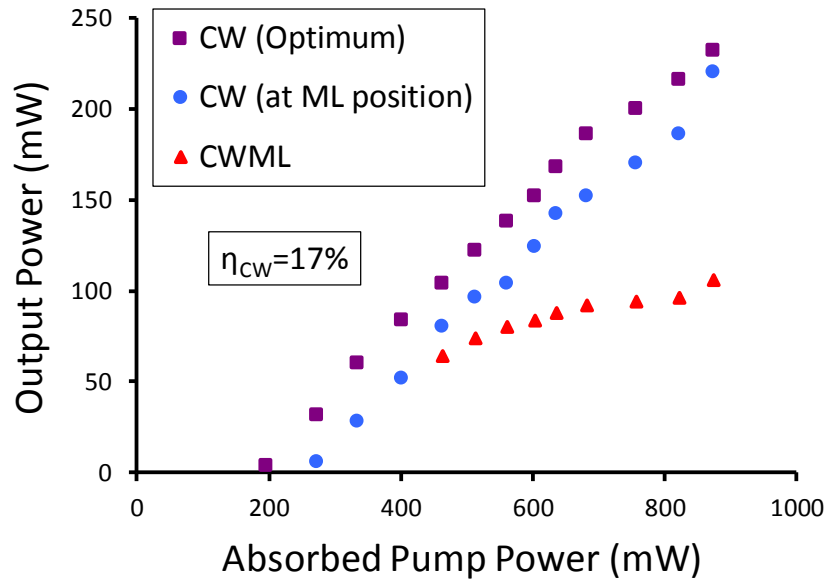


Figure 3. 5 Measured variation of the Cr:LiSAF laser output power as a function of the incident pump power. The data were taken using the tapered diode laser as the pump source at a crystal holder temperature of 15 °C.

During the measurements, the crystal holder temperature was kept at 15 °C to minimize thermal effects. In Fig. 3.6, the measured cw output power of Cr:LiSAF laser employing GMOC, as a function of crystal holder temperature is presented. Note that, when the crystal holder temperature exceeds 20 °C, a sharp decrease in the output power is observed due to thermal

effects. This nonlinear behavior is attributed to lifetime quenching [118] as a result of local temperature levels ($\sim 70^\circ\text{C}$) reached inside the Cr:LiSAF crystal. At elevated temperatures the peak of the Cr:LiSAF gain profile shifted to longer wavelengths. However, this was not a limiting factor for the generated pulsewidths that we obtained. Note also that decreasing temperature of the Cr:LiSAF crystal below 15°C , improves the laser performance by 10–20%. In the experiments, the Cr:LiSAF laser operated at 15°C which is close to the ambient air temperature.

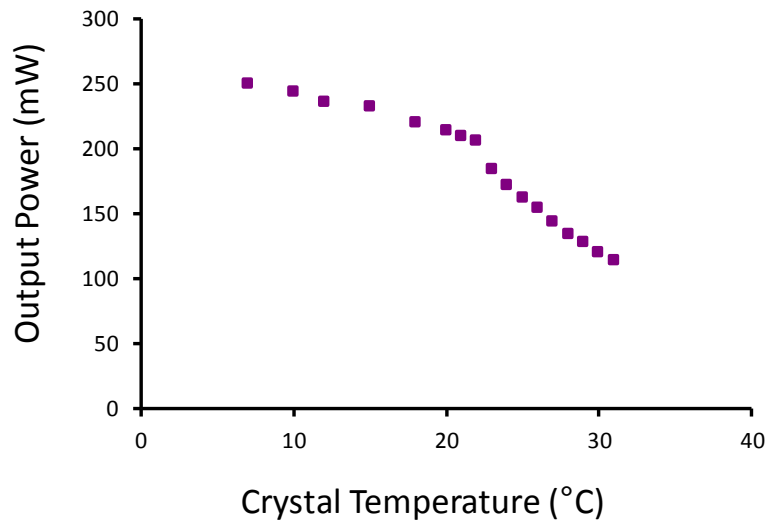


Figure 3. 6 Measured variation of the Cr:LiSAF laser output power as a function of the crystal holder temperature. The data were taken using the tapered diode laser as the pump source at the full-pump power of 1W.

In the previous studies, TD pumped cw Cr:LiSAF laser with a 0.5% output coupler resulted in lasing thresholds as low as 45 mW and slope efficiencies as high as 47% [148]. One of the reasons of this modest performance we obtained in this study was the use of dispersion optimized broadband DCM pairs which had relatively higher passive losses compared to cw laser optimized high reflectors. Also, insertion loss of the fused silica prism pair might have contributed to the decrease in the cw laser performance. Moreover, the effective output coupling of the resonator (1.5%, due to two bounces on the GMOC) was slightly above the optimum value for the Cr:LiSAF lasers operating in similar pumping levels. Increased thermal effects limit the obtainable power levels at high output coupling due to Auger transfer up-conversion process [148]. Cr:LiSAF oscillators have optimum output coupling values depending on several

factors such as available pump power. Higher output coupling of the resonator limits the obtainable average output powers due to above mentioned thermal effects. On the other hand, lower output coupling limits the obtainable peak power levels due to intracavity nonlinearities. In Figure 3.5, CW (at ML position) data shows the measured cw performance at the KLM operation point. Slight decrease of the cw power from 232 to 220 mW indicates a good mode-matching between the pump and laser beams.

The fine tuning of the cavity dispersion was managed by a fused silica prism pair with a separation of 45 cm. By varying the insertion introduced by the second fused silica prism, we could obtain the tuning of the pulsewidths in the 15–100 fs range. Although we could initiate KLM with regular output couplers, with transmission values ranging from 0.1% to 1%, the KLM operation was less stable compared to the GMOC. In this case, obtained pulse widths were limited to around 25 fs. Furthermore, the KLM action disappeared and could not be initiated when dispersion is set for shorter pulses.

The measured variation in the optical spectrum and pulsewidth values as a function of varying cavity dispersion are shown in Fig. 3.7. Here, ΔD is the change in dispersion calculated from the measured prism material insertion difference. For this reason, the specified dispersion values in the figure indicate only the change in dispersion with respect to the absolute dispersion value of the resonator for the shortest pulses. For the ΔD values from $+50 \text{ fs}^2$ to -400 fs^2 (a total cavity dispersion variation of 450 fs^2), the resonator generated femtosecond pulses. Beyond this range, the laser operated in cw regime and fs pulsing could not be initiated. As seen from the general trend of Fig. 3.7, when the dispersion of the cavity was made more negative, longer pulses with narrower spectra were obtained as expected.

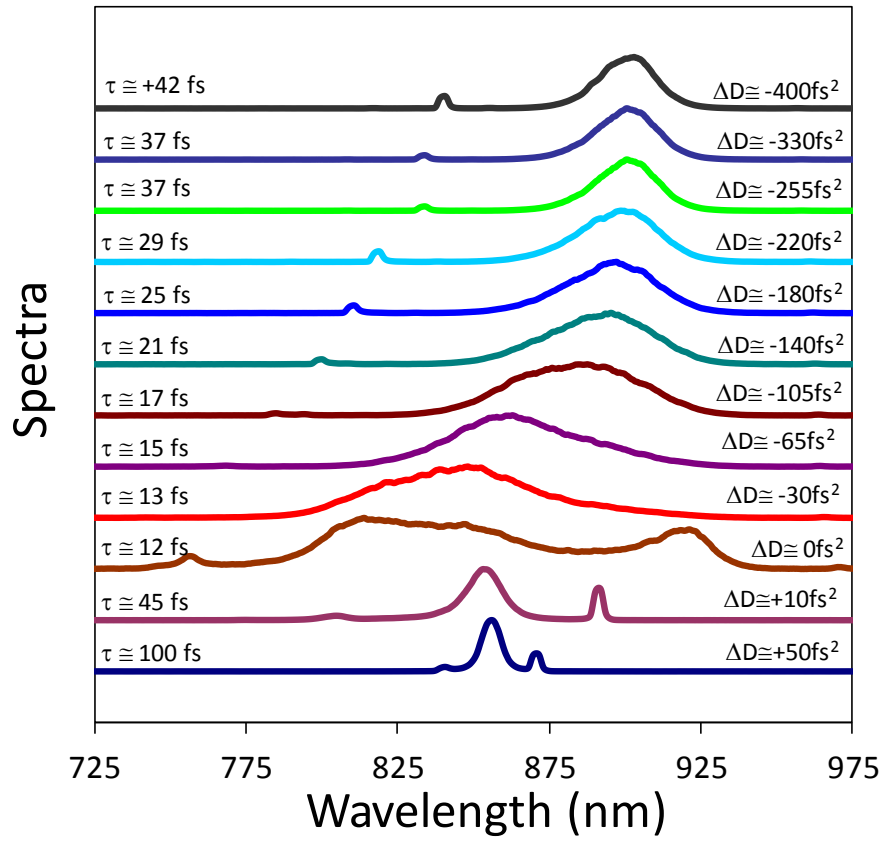


Figure 3.7 Measured variation of the Cr:LiSAF laser output spectrum with varying intracavity dispersion. τ refers to the calculated Fourier transform limited pulsewidth for each optical spectrum. ΔD is the calculated change in the dispersion based on the measured prism material insertion. Here, only the variation of dispersion with respect to the dispersion of the cavity for the shortest pulses is indicated. Hence, specified dispersion values are not absolute. We estimate the absolute dispersion value to be around -50 fs^2 for the shortest demonstrated pulses (12 fs).

Furthermore, the central wavelength of the pulses shifted to longer wavelengths which could be attributed to a slight misalignment in the prism pair or to a shift of the dispersion minimum as the prism insertion was varied. For positive values of ΔD , the laser produced fs pulses with longer pulsewidths (where one actually expects to get shorter pulses). This, we believe, might be due to the presence of local minima within the dispersion bandwidth. Probably, the dispersion and reflectivity profile of the cavity was not optimum to support even shorter pulses. Hence, the optical spectrum sits into one of the dispersion minima resulting in longer pulse generation.

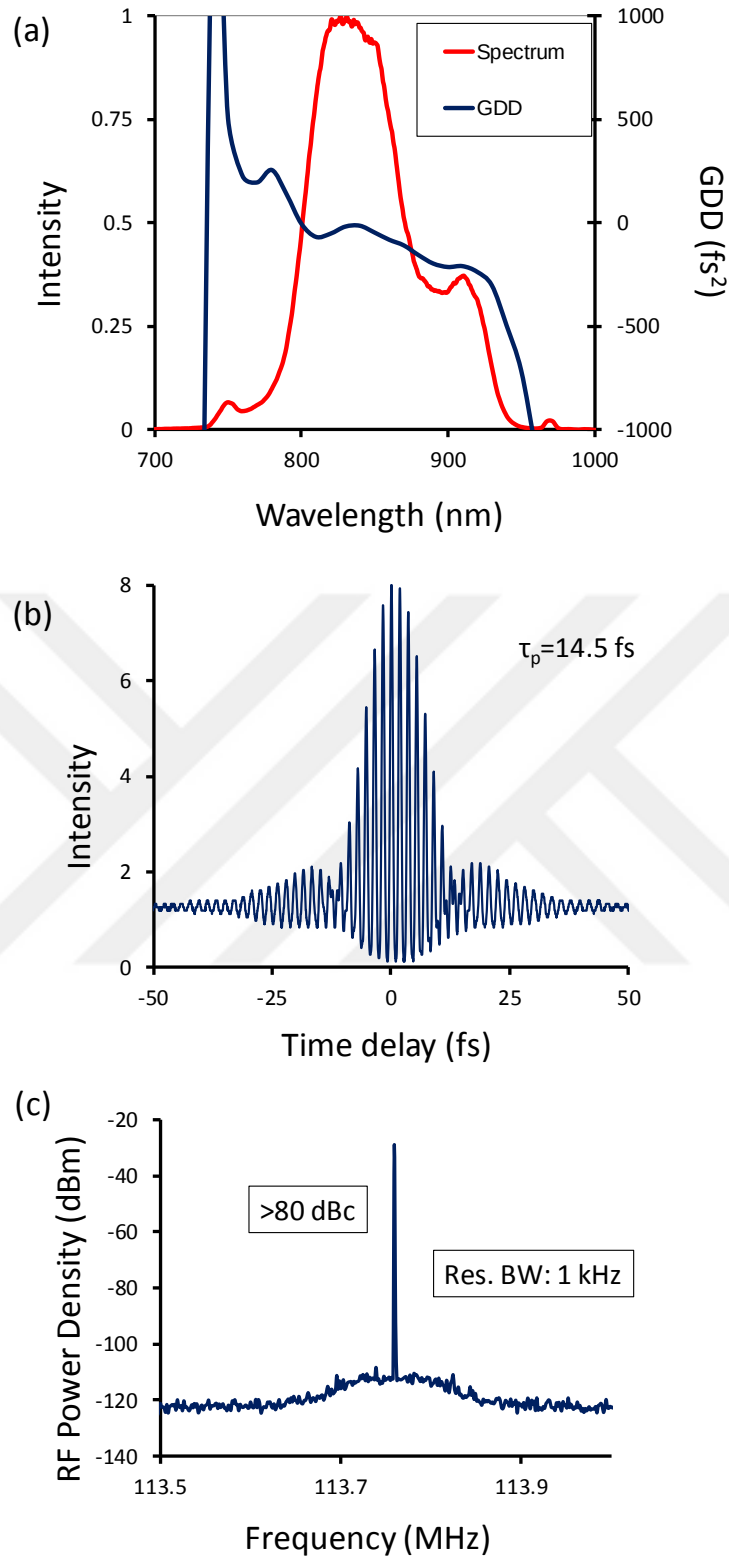


Figure 3. 8 Measured (a) optical spectrum, (b) interferometric autocorrelation, and (c) radio frequency spectrum for the 14-fs long pulses with 106 mW average power. The RF data were taken at 1 kHz resolution.

Figure 3.8 summarizes the mode locking results obtained from the TD pumped Cr:LiSAF laser. Figure 3.8(a) shows the measured optical spectrum which is broad enough (from 740 to 950 nm) to support 12.5-fs pulses. We calculated the minimum achievable time bandwidth product (0.36) and autocorrelation deconvolution factor (1.3) for this spectral shape and measured the pulsewidth as 14.5-fs. The pulses were slightly chirped with a time-bandwidth product of 0.42. The average output power of the laser during mode locked operation was measured to be 106 mW at an absorbed pump power of 850 mW which corresponded to a pulse energy of 940 pJ. The peak power of 60 kW, to the best of our knowledge, represented the highest peak power obtained from any diode pumped Cr:LiSAF oscillator. Our carrier frequency had at least 80 dBc contrast with respect to noise level at 113 MHz. Furthermore, the resonator had an optical-to-optical conversion efficiency of 11%.

Although we used a TD as the pump source which had an asymmetric beam profile with M^2 values of 5 and 1.1 in the slow and fast axes, respectively, we could obtain symmetric beam profiles with almost diffraction limited focusing properties from the Cr:LiSAF laser both in cw and cwml regimes as shown in Figure 3.9. The M^2 values of 1.09 and 1.03 were measured for the cw case in the y and z axis, respectively. During the cw mode-locking operation, M^2 values of 1.08 and 1.01 were measured for the y and z axis, respectively.

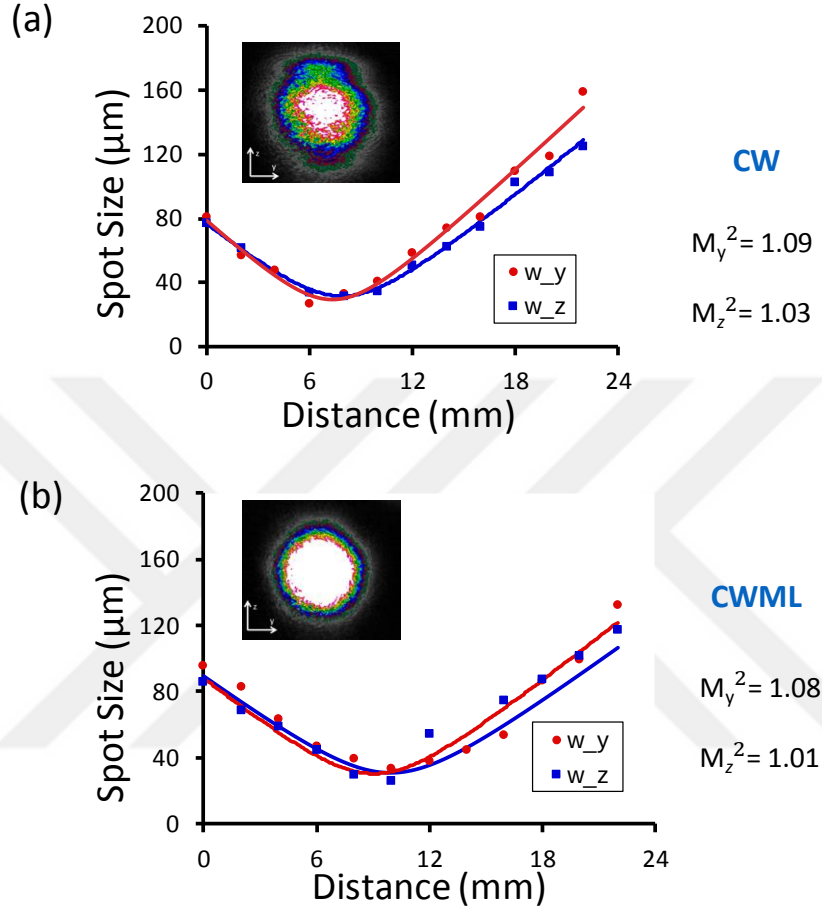


Figure 3. 9 Measured output beam profiles of the tapered-diode pumped Cr:LiSAF laser for the (a) cw and (b) cw mode-locked (cwml) cases. Measured variation of the spot sizes in both axes, for both the cw and cwml cases are also shown. An empirical fit to the measured spot size revealed that M^2 values of the output beams are less than 1.1 in all cases.

Many applications such as multiphoton microscopy, time-resolved photoluminescence, or pump–probe spectroscopy require broad tunability in the mode-locked regime. For this reason, we have also investigated the tuning capability of our KLM Cr:LiSAF laser in the mode-locked regime. An adjustable slit after the fused silica prism pair was used to tune the central wavelength. For each central wavelength, we optimized the dispersion of the cavity for the generation of the shortest pulses. The continuous tuning range of the center wavelength (from 807 to 919 nm) could be obtained as shown in Fig. 3.10.

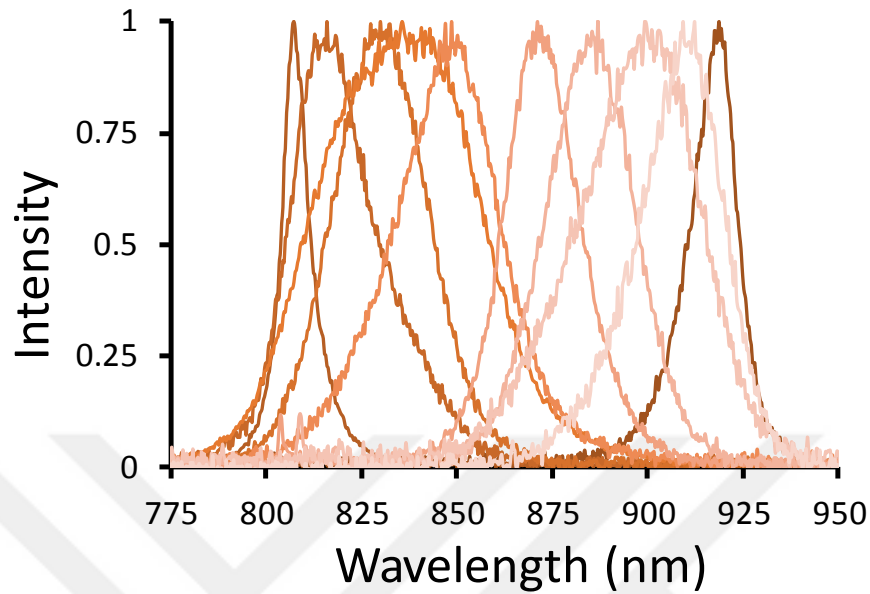


Figure 3.10 Typical spectra from the Cr:LiSAF laser showing tunability of the central wavelength from 807 nm to 919 nm (112 nm bandwidth), for sub-50-fs pulses. The data were taken at the full-pump power of 1 W.

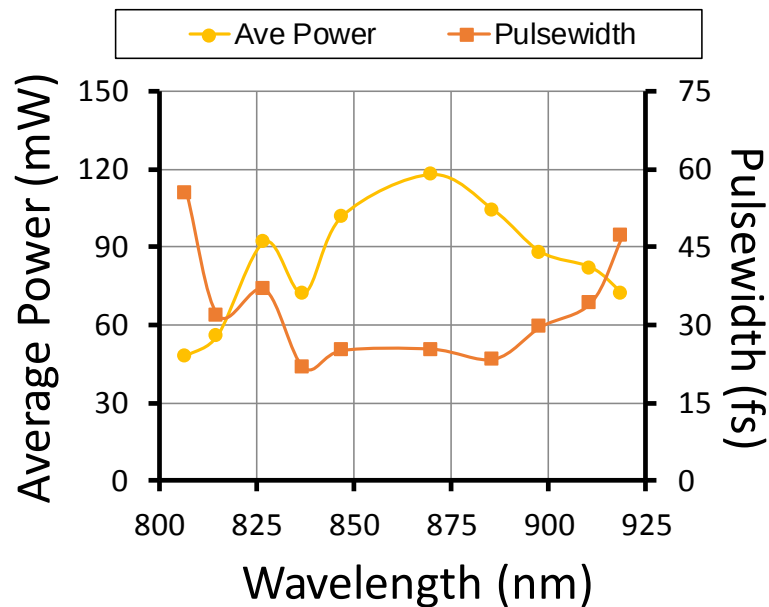


Figure 3.11 Variation of laser output average power and pulsewidth as a function of the central wavelength of the femtosecond pulses. The data were taken at the full-pump power of 1 W.

Figure 3.11 shows the average pulse duration (~ 33 fs) and pulse energies (~ 750 pJ) across the tuning range. In previous studies, femtosecond tuning ranges of 835–910 nm [149] and 809–910 nm [141] were reported for KLM Cr:LiSAF lasers. The single-walled carbon nanotube modelocked Cr:LiSAF laser yielded a fs tuning range from 868 to 882 nm [150]. A fs tuning range from 800 nm to 905 nm could be obtained from an oxidized broadband SBR mode-locked Cr:LiSAF laser [151]. In this study, we could slightly extend the previous results into the infrared region [151].

In summary, a low cost and efficient Kerr-lens mode-locked Cr:LiSAF laser system employing a gain-matched output coupler was presented. 1-W tapered diode laser around 680 nm ($M_x^2 = 1.1$, $M_y^2 = 5$, $B = 450$ mW/ μm^2) was used as a pump source. Stable and robust KLM operation was sustained by the use of GMOC mirror. Sub 15 fs pulses with peak powers as high as 60 kW was generated. We extended the femtosecond tuning range from 807 to 919 nm. To the best of our knowledge, we obtained the highest peak powers and the broadest fs tuning range obtained from Cr:LiSAF laser oscillators to date. This study clearly demonstrates that GMOC mirror reduces the stringent requirements of Kerr lens mode locking even in gain media with a low n_2 like Cr:LiSAF. The use of GMOC significantly improves the long-term stability as well as the efficiency of the laser. Our experimental results in this study were limited by the reflectivity and dispersion bandwidth of the available optics. With better optimized optics, we expect the Cr:LiSAF lasers using the GMOC technology to enable the generation of pulsewidths down to 7-fs, with broader tuning ranges (800–1000 nm) , and higher peak powers (above 100 kW).

Chapter 4

Sub-100-fs Pulse Generation from a Kerr-lens mode-locked multipass-cavity Alexandrite laser

We start this chapter with a brief overview of Alexandrite gain medium properties. Then we focus on the generation of the shortest femtosecond pulses, reported to date, from a Kerr-lens mode-locked multipass cavity (MPC) Alexandrite laser around 750 nm. In the experiments, we investigated the performance of both the short and extended resonators where the Alexandrite gain medium was pumped with a continuous-wave, 532-nm laser. Furthermore, we explain how the use of an extended cavity eliminated the multi-wavelength spectral instabilities observed during the continuous-wave operation of the short cavity. The Kerr-lens mode-locked MPC Alexandrite laser produced nearly transform-limited, 70-fs pulses at a pulse repetition rate of 5.6 MHz with only 1W of pump power. The time-bandwidth product was further measured to be 0.331.

4.1 Alexandrite Gain Medium Properties

Alexandrite (Cr^{3+} -doped Chrysoberyl, $\text{Cr}^{3+}:\text{BeAl}_2\text{O}_4$) is the first tunable solid state laser operated at room temperature [40]. It is the third gain medium after Ruby ($\text{Cr}^{3+}:\text{Al}_2\text{O}_3$) [33] and $\text{Cr}^{3+}:\text{YAG}$ [152], in which Cr^{3+} -ion was reported to lase. In 1963, possibility of the laser operation of the R-line in Alexandrite was first suggested by Farrel and Fang [153]. The first demonstration of lasing in Alexandrite was reported by Morris and Cline in 1974 [154]. In 1977, Jenssen and Walling discovered that lasing occurs in the vibronic sideband as well as the expected strong R-line and reported wavelength tuning, temperature dependent behavior and Q-switched operation of the Alexandrite laser [40, 155, 156]. Before the vibronic operation of the Alexandrite, the only commercial solid-state lasers were the fixed wavelength ruby and Nd: YAG lasers. Alexandrite can operate both as a four-level vibronic laser

and as a three-level laser system. In the latter case, it has a high threshold, fixed output wavelength (680.4 nm at room temperature), and relatively low efficiency. Here, we are interested in the vibronic operation of the Alexandrite gain medium. The simplified energy diagram of the four-level Alexandrite laser can be described as in the figure below.

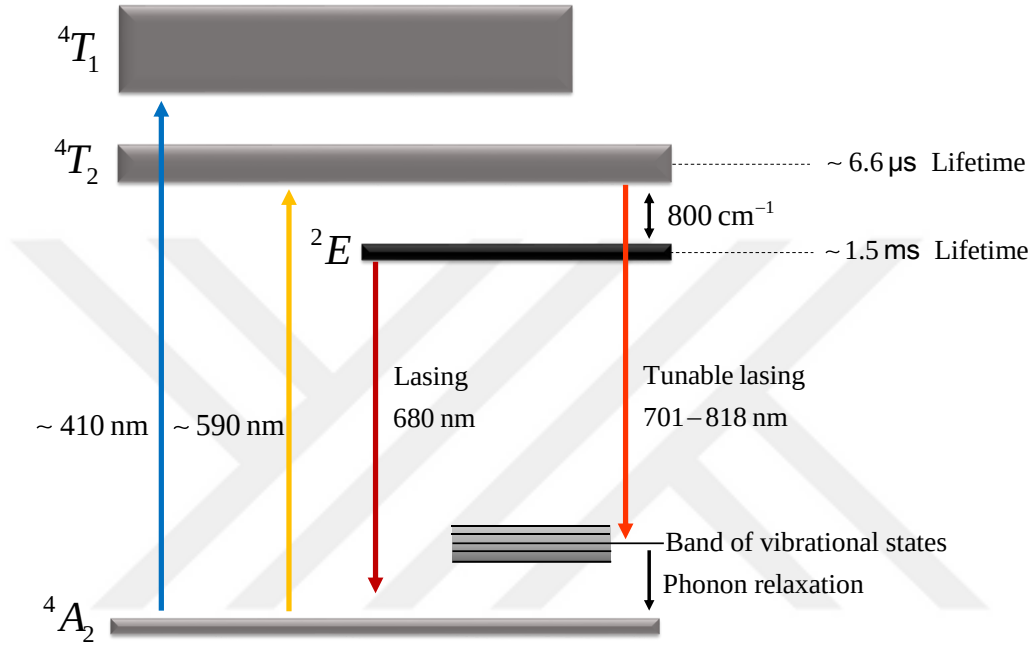


Figure 4. 1 Simplified 5-level energy bands for Alexandrite at 4K (adopted from [156, 157])

Apart from the storage level $2E$, which is 800 cm^{-1} below the energy level $4T_2$, this model is similar to the classical four-level laser model. Alexandrite possesses two strong and broad absorption bands with absorption peaks near 410 nm and 590 nm. The pump laser populates one of the vibrational sublevels of the upper levels which then rapidly depopulates in the nonradiative process into the $2E$ level. The intrinsic lifetime of the $4T_2$ state is around $6.6 \mu\text{s}$, while the $2E$ state has a lifetime of around 1.54 ms. We note again that the dynamics of the $4T_2$ level cannot be separated from that of the metastable $2E$ level, which acts as a reservoir for the ions of $4T_2$ level. Laser transitions from the storage level to the ground state $4A_2$ is analogous to the operation of ruby lasers. The four-level, vibronic lasing stems from the emission from the $4T_2$ state to excited vibronic states within the $4A_2$ level [40, 158, 159].

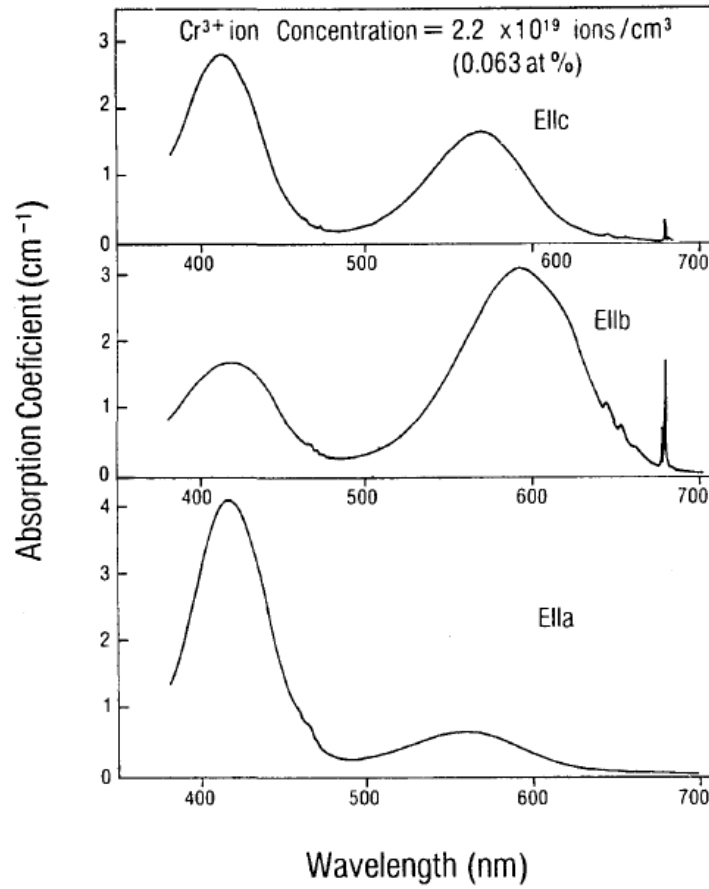


Figure 4. 2 Absorption spectrum of Alexandrite [40].

Over the years, the absorption band of the Alexandrite shown in Fig. 4.2 was exploited by flash lamps, arc lamps, diodes and LEDs as the pump sources of the Alexandrite lasers [160-172]. Also, diffraction limited green pump sources were presented as an alternative approach to improve output power and the beam quality of the Alexandrite lasers [173, 174]. Before the advent of the Ti:sapphire laser, Alexandrite was the most commercially available tunable laser because of its favorable thermo-mechanical properties. Alexandrite has a high fracture resistance which is higher by a factor of 3-5 compared to Nd:YAG. Alexandrite has a low stimulated emission cross-section ($0.7 \times 10^{-20} \text{ cm}^2$) which requires high laser fluence for efficient gain extraction. However, this drawback is compensated with the Alexandrite's high optical damage threshold ($>270 \text{ J cm}^{-2}$). The thermal conductivity of Alexandrite is very close to that of Ti:sapphire. Furthermore, emission cross section-fluorescence lifetime ($\sigma_{em} \tau$) is

larger compared to Ti:sapphire as shown in Table 4.1. This, in principle, allows the operation of Alexandrite laser with lower threshold pump powers.

Table 4. 1 Comparison of the spectroscopic and laser parameters of Alexandrite and Ti:sapphire.

Gain medium	Alexandrite ($\text{Cr}^{3+}:\text{BeAl}_2\text{O}_4$)	Ti:sapphire ($\text{Ti}^{3+}:\text{Al}_2\text{O}_3$)
Tuning Range [nm]	701-858 [156, 175]	660-1180 [176]
Upper-state room temperature lifetime [μs]	260 [177]	3.2[45]
Thermal conductivity [W/K-m]	23 [177]	28 [112]
$\sigma_{\text{em}}[\times 10^{-20} \text{ cm}^2]$	0.7 [177]	41[45]
Refractive index	1.74[156]	1.76[111]
Nonlinear refractive index coefficient [m^2/W]	$2 \pm 0.3 \times 10^{-20}$ [177]	3.2×10^{-20} [109]
$\sigma_{\text{em}}\tau[\times 10^{-20} \text{ cm}^2\text{-}\mu\text{s}]$	183 [177]	131 [45]
Crystal figure of merit (FOM)	3000 [160]	150 [90]
Intrinsic slope efficiency	65 [170]	64 [114]

In addition to its favorable thermo-mechanical properties and broad tunability, Alexandrite lasers can be utilized in low-cost, compact and mobile systems since they can be pumped with well-developed red diode lasers. Thus, Alexandrite gain medium is suitable for the development of tunable, high-power, diode-pumped lasers in the near-infrared [178-182]. Furthermore, by taking the harmonics of the Alexandrite laser, a tunable source in the ultraviolet spectral range can be obtained. Alexandrite lasers operating in the 700-858 nm wavelength range [156, 175, 183] have been used in numerous applications. Clinical dermatology is one of the fields that utilize Alexandrite lasers [184-186]. In the space-airborne remote sensing and lidar systems, Alexandrite lasers are preferred due to their strong thermo-mechanical properties, tunability and mobility [168, 171, 187-191]. Although diode-pumped Nd:YAG/Nd:YVO₄ lasers at 1064 nm are available for similar applications, they do not provide tunability. Optical parametric conversion brings high complexity, loss of efficiency and increased cost to the solution of this problem. Furthermore, the harmonics of the Nd-based systems (532nm/355nm/266nm) miss the entire wavelength regions. Moreover, the peak emission of Alexandrite (~750 nm) is important for optimum excitation of several widely used fluorophores such as 4'-6-diamidino-2-phenylindole dihydrochloride (DAPI), Alexa fluor 488, indo-1, and fura-2 in multiphoton microscopy [17, 22, 192-196].

In particular, Munk *et al.* developed mobile lidar systems which can operate even under harsh environmental conditions e.g. polar conditions in the Antarctic for atmospheric measurements [168,

189]. Damzen *et al.* also made important contributions to the improvement of Alexandrite lasers for the satellite-based remote sensing systems. They could obtain more than 26W output power from a diode-pumped Alexandrite laser [178]. As stated previously, Alexandrite's application fields can be extended by high harmonics generation. In this regard, Imai *et al.* studied on the second and third harmonics generation capability of Q-switched Alexandrite lasers [197, 198]. However, despite being heavily used in continuous-wave (cw) [174, 199-201] and picosecond pulsed regime [202-205], until recently femtosecond pulse generation capability of Alexandrite was not utilized. Ghanbari *et al.* reported 170-fs pulses from a Kerr-lens mode-locked Alexandrite laser with a pump power of 8 W at 532 nm [206]. The same group generated 380-fs pulses from Alexandrite laser by using a quantum dot semiconductor saturable absorber mirror [207].

Here, we report to the best of our knowledge, the shortest femtosecond pulses generated from a Kerr-lens mode-locked Alexandrite laser. The resonator produced 70-fs nearly transform-limited pulses at the center wavelength of 750 nm. In the experiments, the optical path length of the resonator was increased by using a multipass-cavity (MPC) extension to increase the output pulse energy and to eliminate the Q-switching instabilities. The MPC extension further enabled the generation of femtosecond pulses with reduced thermal effects since mode locking could be achieved with only 1W of pump power at 532 nm.

4.2 Experimental Details

Figure 4.3(a) shows the multipass-cavity (MPC) Alexandrite laser resonator which was used for the femtosecond pulse generation experiments. The continuous-wave pump beam (532 nm OPUS, Laser Quantum) was focused to an estimated beam waist of 23 μm inside the 10-mm-long Brewster-cut Alexandrite gain medium by using a converging lens (L , $f=75$ mm). The absorption of the Alexandrite crystal was measured to be 93% at the pump wavelength of 532 nm. During the experiments, the gain medium was surrounded by an indium foil and kept near 20 °C by using a water-cooled, copper holder. Initially, an x-cavity was built by using one flat (M6) and two curved (M1 and M2, $R=75$ mm), high reflectors in addition to a flat output coupler (OC).

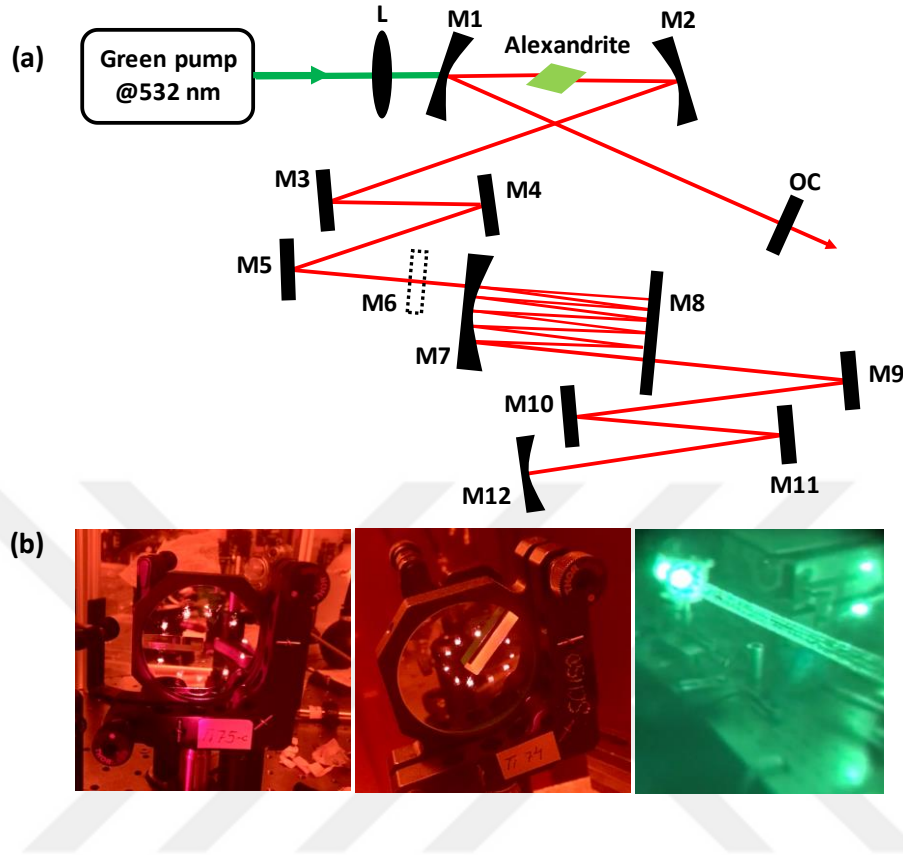


Figure 4.3 (a) Experimental setup of the 532-nm pumped, multipass-cavity Alexandrite laser. (b) Photographs of the curved (left) and flat (middle) multipass-cavity mirrors, and the circulating beam inside the multipass-cavity (right).

To increase the optical path length of the resonator, the high reflector arm of the x-cavity was further extended by introducing a multipass-cavity (MPC) as shown in Fig. 4.3(a). Furthermore, three flat high reflectors (M3, M4, and M5) were added to the resonator and used as steering mirrors to adjust the inclination angle of the beam entering the MPC. The curved (M7, ROC=2 m) and the flat (M8) high reflectors of the MPC were separated by 114 cm [see Fig. 4.3(a)]. Both high reflectors (M7 and M8) had rectangular notches to allow the entry and exit of the circulating beam within the MPC. Figure 4.3(b) shows the photographs of the MPC mirrors [M7 (left) and M8 (middle)] and the circulating beam (right) inside the MPC. As can be seen from Fig. 4.3(b), 10 roundtrips were completed before the beam exited the high reflector M8. The separation (114 cm) between M7 and M8 was adjusted to make the MPC q-preserving with $n=11$ and $m=6$. Here, n and m correspond to the number of full roundtrips that must be completed and the number of semicircular arcs that the beam completes before leaving the MPC. Note

that as can be seen from Fig. 4.3(b), the circulating beam cannot complete 11 full roundtrips due to the presence of the notches. The missing roundtrip was added by including an additional curved high reflector (M12) with a radius of 1 m, located at a distance of 114 cm from M8 [85]. The resulting MPC configuration was q-preserving and provided an additional roundtrip optical path length of 50.2 m. Here, q refers to the complex beam parameter of the Gaussian beam and the term “q-preserving” indicates that in this configuration, the beam returning from the MPC has the same spot size and radius of curvature as the incident beam. The overall pulse repetition rate of the resonator was hence reduced from 107 to 5.6 MHz, corresponding to a nearly 20 times increase in the output energy of the mode-locked laser operated with the same output coupler and at the same average output power. This further enabled the mode-locked operation of the Alexandrite laser with reasonably low pump power at 532 nm. To obtain femtosecond pulses from the Alexandrite laser, the intracavity dispersion of the composite resonator was managed by using dispersion control mirrors. The mirrors M4, M5, M7, M8, M9, and M12 had a negative group delay dispersion (GDD) of -50 fs^2 per bounce. In addition, the mirrors M10 and M11 had a negative GDD of -100 fs^2 and the mirror M3 had a negative GDD of -40 fs^2 per bounce. By taking into account the dispersion due to the gain medium (60 fs^2 per mm [75]), air (20 fs^2 per m), the mirrors M1, M2, and OC ($\sim 5 \text{ fs}^2$ per bounce), the net GDD of the composite resonator shown in Fig. 4.3(a) was estimated to be around -533 fs^2 near 750 nm.

4.3 Continuous-wave Operation Results

Before mode-locking experiments, we investigated the continuous-wave power performance of the Alexandrite laser for the short and extended resonators. Figure 4.4(a) shows the measured power efficiency of the x-cavity Alexandrite laser by using four output couplers with 0.1%, 0.25%, 0.5%, and 1.7% transmission near 750 nm. The measured slope efficiencies with four different output couplers were 5.1%, 9.7%, 19.2%, and 23.2%, respectively. The roundtrip passive loss of the x-cavity Alexandrite laser was estimated to be 0.48% by using the threshold pump power data shown in Fig. 4.4(b).

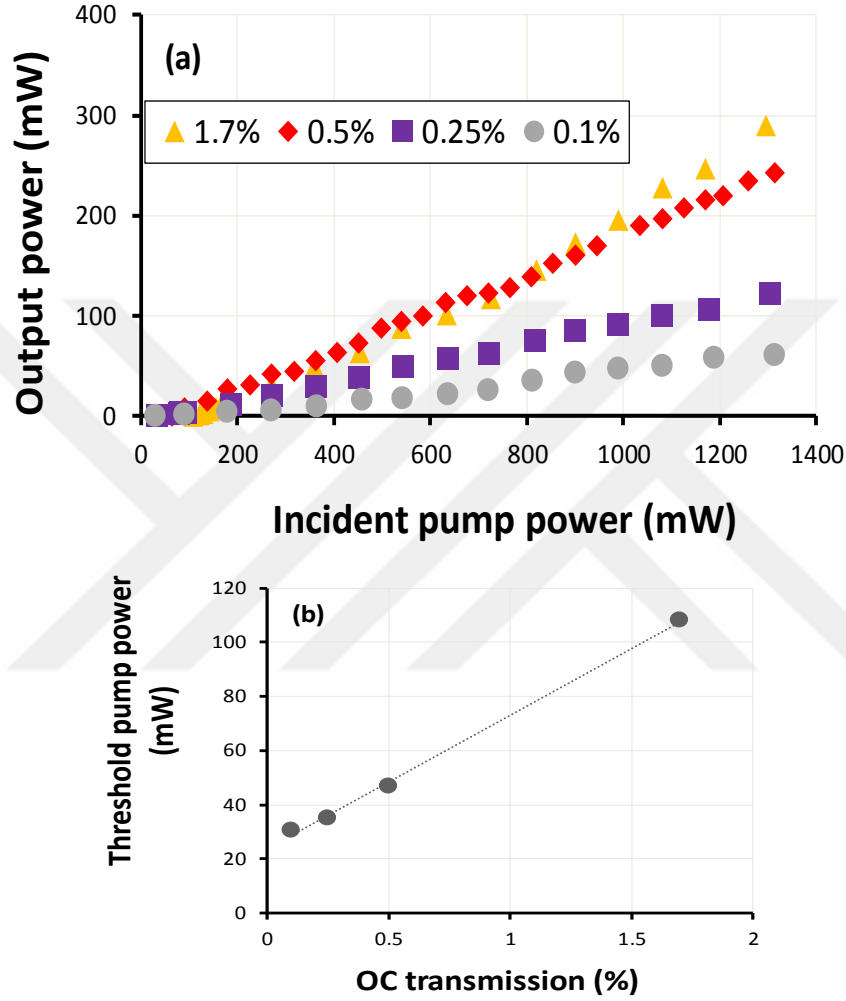


Figure 4. 4 (a) The continuous-wave power efficiency data of the x-cavity Alexandrite laser measured with four output couplers. **(b)** The measured threshold pump power with respect to the output coupler transmission.

Figure 4.5 shows the continuous-wave power efficiency data of the short- and extended-cavity Alexandrite laser operating with the 0.25% output coupler. The MPC extension decreased the power slope efficiency of the x-cavity Alexandrite laser from 9.7% to 2.8%. The roundtrip passive loss introduced by the MPC was further estimated to be 1.8% by using the passive loss of the x-cavity and the measured threshold pump powers shown in Fig. 4.5. In the measurements, it was necessary to

minimize thermal lensing induced power fluctuations of the output by optimizing the position of the curved mirrors (M1 and M2).

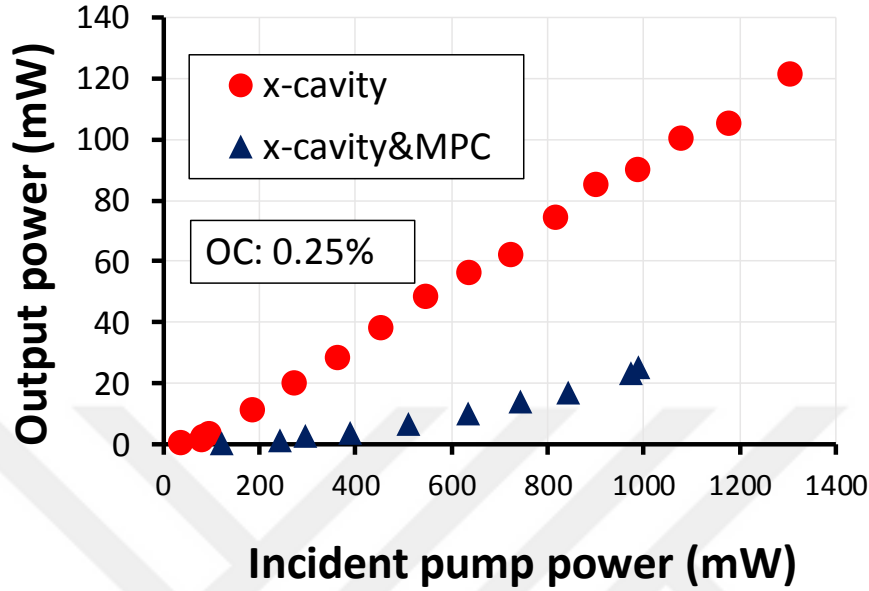


Figure 4. 5 The measured power efficiency of the short- and multipass-cavity (MPC) Alexandrite laser operating with the 0.25% output coupler.

Figure 4.6 (a) shows the measured output spectrum of the continuous-wave Alexandrite laser without the MPC extension. As can be seen, the spectrum break-up occurred at multiple wavelengths and it was not possible to operate the laser with a stable spectral output. In this configuration, mode-locked operation free of Q-switching instabilities could not be observed. This problem was overcome by going to the extended-resonator configuration where a stable continuous-wave spectrum could be easily obtained [Fig. 4.6(b)]. Q-switching instabilities were further eliminated in the extended-cavity configuration due to the higher available pulse energies. Multipass cavity extension increased the intracavity pulse energy and provided an enhanced Kerr nonlinearity which eliminated the Q-switching instabilities during mode-locked operation. These observations are in qualitative agreement with previous simulations in which the chaotic behavior in Alexandrite lasers was shown to depend on the cavity length [208].

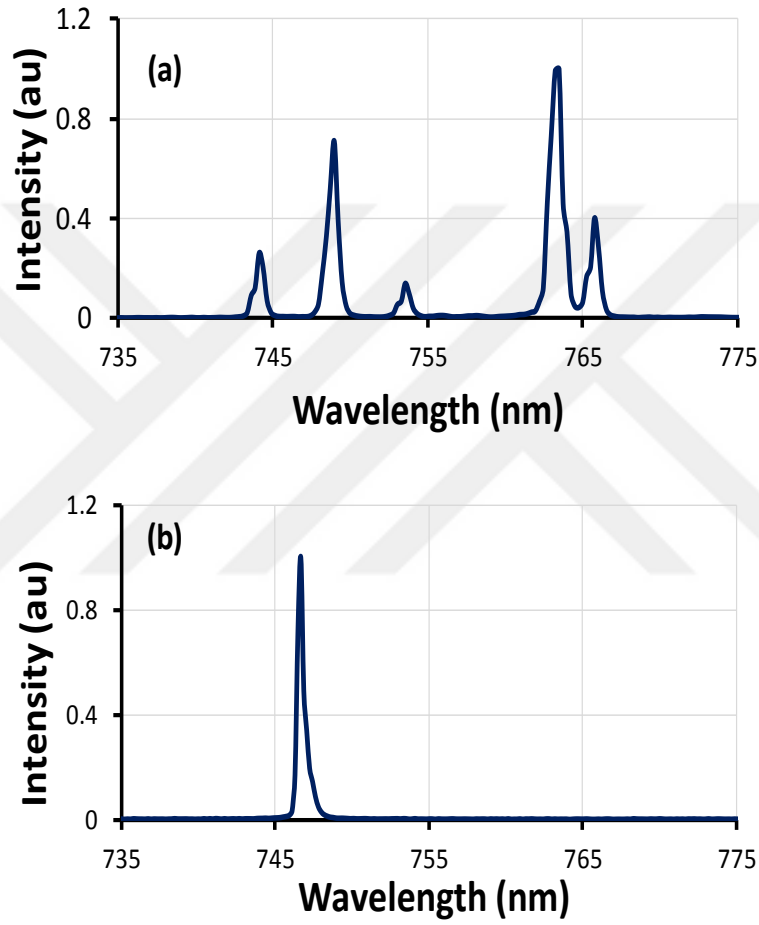


Figure 4.6 The measured continuous-wave output spectrum of **(a)** the short-cavity and **(b)** the extended-cavity Alexandrite laser.

4.4 Mode-locked Operation Results

When the Q-switching instabilities were eliminated, the Kerr-lens mode-locked (KLM) operation of the Alexandrite laser could be achieved. Figures 4.7 and 4.8 summarize the mode locking results obtained

from the Kerr-lens mode-locked operation of the Alexandrite laser. Figures 4.7 (a) and (b) show the measured output spectrum and the interferometric autocorrelation trace of the KLM Alexandrite laser. The measured optical spectrum and interferometric autocorrelation trace yielded a spectral width of 9 nm and a temporal duration of 70 fs for the generated pulses. The corresponding time-bandwidth product of the pulses was estimated to be 0.331 by assuming a sech^2 intensity profile. During mode-locking operation, the output power was around 4 mW, corresponding to 0.7 nJ of pulse energy and 10 kW of pulse peak power at a pump power of 1W. Figures 4.8(a) and (b) further show the measured radio-frequency (RF) spectrum and the pulse train of the Kerr-lens mode-locked MPC Alexandrite laser. The measurements shown in the figures indicated that the photodetected RF signal at the pulse repetition of 5.6 MHz was at least 64 dBc above the noise floor and the mode-locked operation was obtained without Q-switching instabilities.

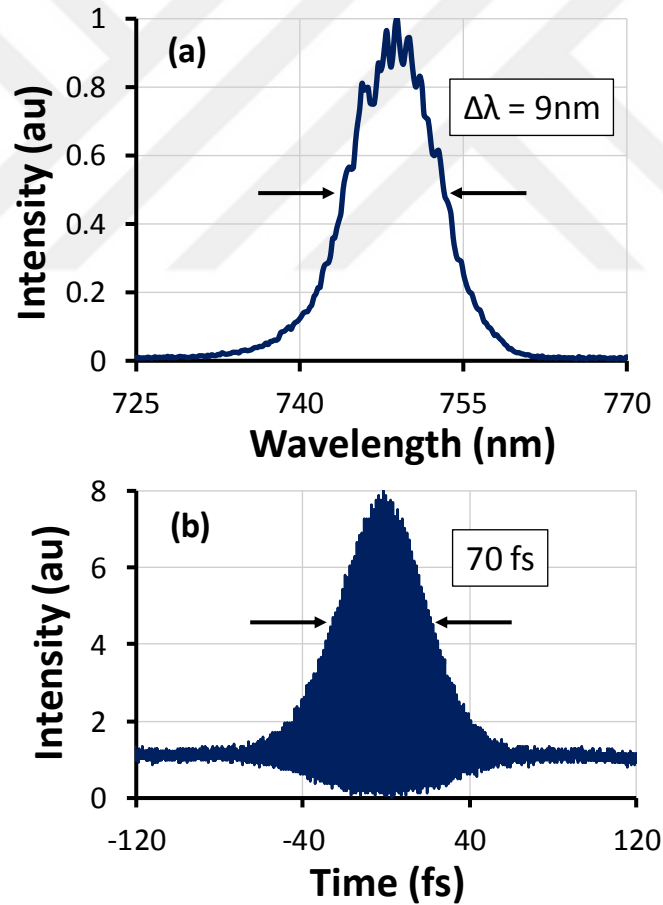


Figure 4. 7 (a) The measured optical spectrum and **(b)** the interferometric autocorrelation trace of the 70-fs pulses obtained from the Kerr-lens mode-locked Alexandrite laser.

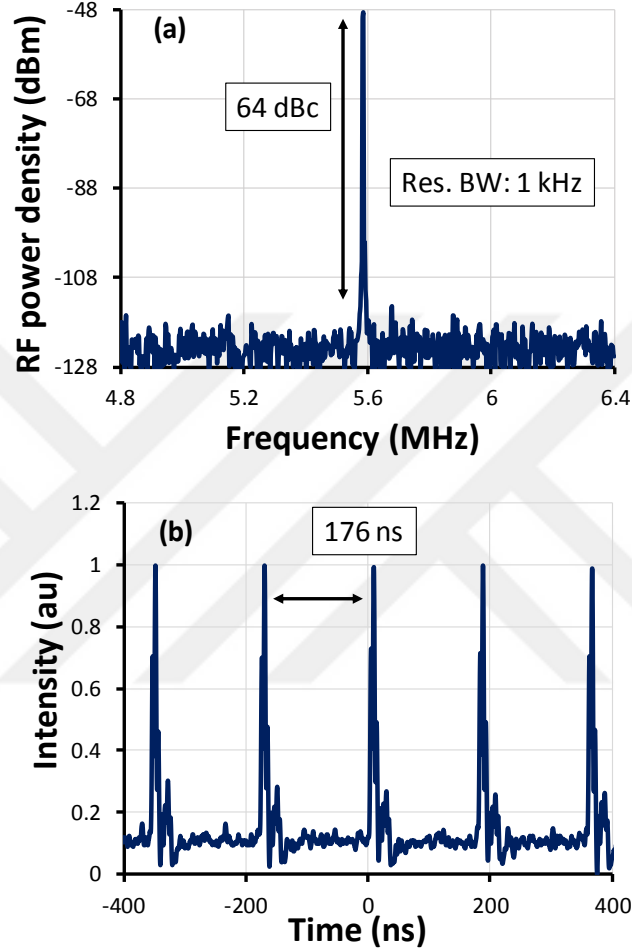


Figure 4. 8 The measured **(a)** radio frequency (RF) spectrum and **(b)** pulse train of the pulses generated with the Kerr-lens mode-locked Alexandrite laser at the repetition rate of 5.6 MHz. During the RF spectrum measurements, the resolution bandwidth (Res. BW) was 1kHz.

In conclusion, we generated the shortest femtosecond pulses from a Kerr-lens mode-locked Alexandrite laser operating near 750 nm. We could eliminate the multi-wavelength spectral instabilities observed during the continuous-wave operation of the short cavity with the use of an extended cavity. The optical path-length increase with the multipass-cavity extension reduced the repetition rate of the Alexandrite laser from 107 to 5.6 MHz. The achieved intracavity pulse energies provided enhanced Kerr nonlinearity and eliminated the Q-switching instabilities. The resonator produced 70-fs transform-

limited pulses with a time-bandwidth product of 0.331. The fundamental beat-note of our carrier frequency had a contrast of 64 dBc with respect to the noise floor at 5.6 MHz. The energy and peak power of the generated pulses were 0.7 nJ and 10 kW, respectively with a pump power of 1W at 532 nm. The tuning bandwidth of Alexandrite gain medium is wide enough to support the generation of sub-10 fs pulses [161]. In our particular case, the estimated dispersion of our resonator was positive at wavelengths below 740 nm. We expect that shorter pulses could be generated with further dispersion optimization.



Chapter 5

Graphene mode-locked femtosecond Alexandrite laser

There are numerous techniques to achieve pulsed operation of lasers. As passive mode-lockers or Q-switchers, the saturable absorber (SA) materials are preferred to achieve pulsed output. This is because they enable pulse generation without the employment of high cost and complex modulators as in the case of active methods. In saturable absorber mode-locking, generated pulse widths are limited by the relaxation times of the carriers, resulting in much shorter pulse durations compared to active mode locking for the same gain bandwidth. Historical evolution of SAs starts nearly with the Maiman's first successful operation of laser. The first SA-based pulse generation was reported in 1964 where dye and coloured glass filter were used to Q-switch a ruby laser [209, 210]. In the beginning of 1990s, semiconductor saturable absorber mirrors (SESAMs) were proposed for pulse generation. However, their limitations such as narrow operating bandwidth and costly fabrication led to search for novel materials. Later, nanomaterials (eg. zero-dimensional quantum dots (QDs), 1D carbon nanotubes (CNTs), 2D graphene) with remarkable physical properties stemming from their reduced dimensionality were used as saturable absorbers [211-214]. There are numerous early studies on graphene [215-221]. For example, Wallace (1947) developed the band theory of the graphite which consists of multi layers of graphene with "tight binding" approximation [222]. In 2004, Andre Geim and Konstantin Novoselov at Manchester University managed to extract graphene by a famous Scotch tape method and they received the Nobel Prize in Physics in 2010 [223]. Their study led to an explosion of research on graphene. Due to its unique properties that will be detailed in the following section, graphene has become a promising material for high frequency electronics and photonics.

The following section starts with a brief discussion of the optical properties of graphene. We then focus on the use of graphene as a saturable absorber for the initiation of femtosecond pulses from the Alexandrite laser. To the best of our knowledge, this study represents the first

use of graphene as a saturable absorber in an Alexandrite laser near 750 nm. This operation wavelength also represents the shortest ultrafast pulse generation wavelength reported to date for graphene. The resonator produced 65-fs nearly transform-limited pulses with a time-bandwidth product of 0.319. In the following sections experimental details and results of graphene mode locked Alexandrite laser will be presented.

5.1 Optical Properties of Graphene as a Saturable Absorber

Graphene is a two-dimensional hexagonal arrangement of Carbon atoms [224]. The corresponding reciprocal lattice is also hexagonal as in the Figure 5.1(a). Band structure calculations further show that graphene has a linear dispersion as sketched as in the Figure 5.1(b). In recent years graphene has attracted great interest in the fields of optoelectronics and photonics [225, 226] due to its exceptional electrical and optical properties such as high carrier mobility ($\sim 200\,000\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$) at room temperatures [227], high thermal conductivity ($5000\text{ W m}^{-1}\text{ K}^{-1}$) [228], high current carrying capacity ($\sim 1.8\text{ A cm}^{-2}$) [229], and high optical damage threshold ($\sim 3 \times 10^{12}\text{ W cm}^{-2}$) [230]. In addition to its superior intrinsic properties, ease of growth and integration with the current CMOS technology [231-235] have made it suitable for the high-frequency electronics [236-243]. Apart from its electrical applications, graphene is an ideal saturable absorber for the ultrafast lasers with its ultrafast carrier dynamics [244-247] and ultrabroad saturable absorption [248-251]. It has been utilized to initiate pulsed operation of a wide range of lasers [252, 253].

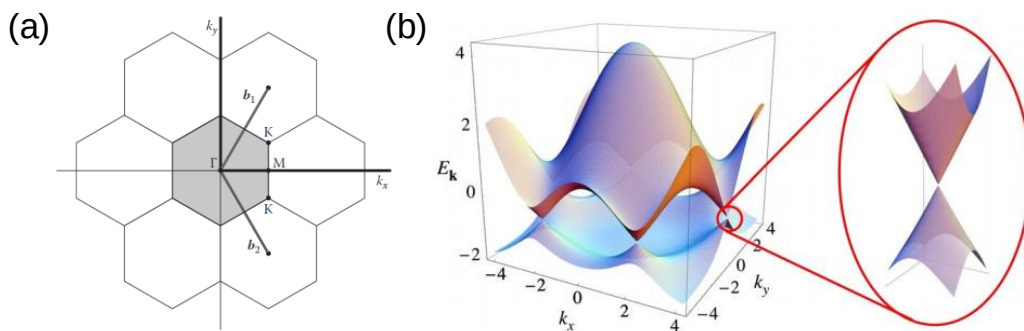


Figure 5.1 (a) Reciprocal lattice of graphene with the reciprocal lattice vectors $\mathbf{b}_1$ and $\mathbf{b}_2$ [254], (b) Electronic dispersion relation (plot of electron energy as a function of wavevector $\mathbf{k}$ in the xy plane) for graphene Inset: the energy bands close to one of the Dirac points [225].

The charge carriers (electrons and holes) of graphene possess ultrafast dynamics resembling relativistic massless Dirac particles. Dawlaty *et al.* measured two distinct time scales associated with the relaxation of photo-generated carriers in epitaxial graphene by using an ultrafast optical pump-probe spectroscopy system. The measurements revealed that the initial fast relaxation transient was in the 70–120 fs range and the following slower relaxation process was in the 0.4–1.7 ps range [252]. The charge carriers in graphene obey a linear dispersion relation given by

$$E = \hbar v_F |\mathbf{k}| \quad (5.1)$$

where $\mathbf{k}$ is the wave-vector, $\hbar$ is the Planck's constant, $v_F \approx 10^6 \text{ ms}^{-1}$ is the Fermi velocity of massless Dirac fermions. This result was first obtained by Wallace [222].

Graphene's band structure, the zero band gap and linear dispersion of Dirac electrons, ensures the existence of electron-hole pair excitations in resonance with any incoming photon from the visible to the far-IR spectral range [244, 255-257]. The interaction between charge carriers and ultrafast optical pulses produces a non-equilibrium carrier population in the valence and conduction bands, which then relaxes on an ultrafast time scale [252]. This band structure offers graphene the advantage of an ultra-broad absorption.

Figure 5.2 shows the broad optical absorption spectra of graphene in different spectral regions. The absorption band of graphene can be seen from visible to mid-infrared (Fig.5.2 (a)), and from visible to terahertz frequency range (Fig.5.2 (b)). Optical absorption of graphene is determined by the optical conductivity $\sigma(\omega)$ which consists of interband $\sigma_{inter}(\omega)$ and intraband $\sigma_{intra}(\omega)$ terms [257-259] given by

$$\sigma_{inter}(\omega) = j \frac{e^2 \omega}{\pi} \int_{\Delta}^{\infty} d\varepsilon \frac{(1 + \Delta^2 / \varepsilon^2)}{(2\varepsilon)^2 - (\hbar\omega + j\Gamma)^2} \times [f(\varepsilon - E_F) - f(-\varepsilon - E_F)] \quad (5.2)$$

$$\sigma_{intra}(\omega) = j \frac{e^2 / \pi \hbar^2}{\omega + j / \tau} \int_{\Delta}^{\infty} d\varepsilon (1 + \Delta^2 / \varepsilon^2) \times [f(\varepsilon - E_F) + f(\varepsilon + E_F)] \quad (5.3)$$

where $f(\varepsilon - E_F)$ is the Fermi distribution function, Γ governs the interband transition, 2Δ is the bandgap, and τ is the momentum relaxation time resulting from carrier intraband scattering.

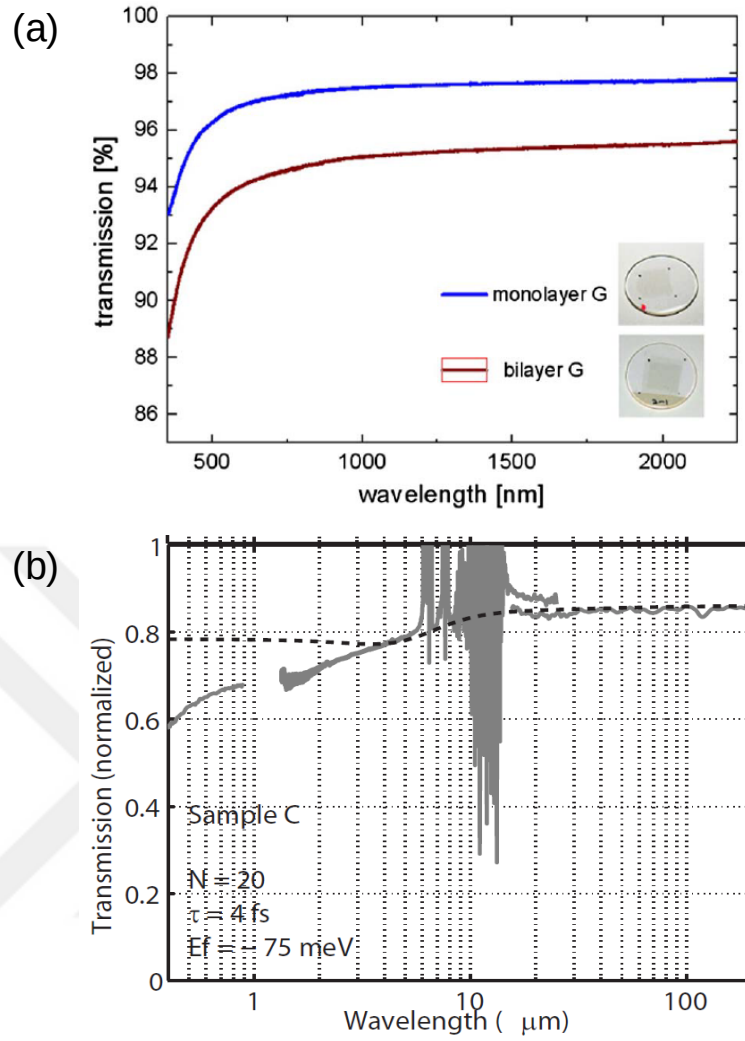


Figure 5. 2 (a) Linear transmission of monolayer and bilayer graphene and photographic images of SAs (inset)[256]. **(b)** Measured transmission spectra (solid gray lines) of sample C from 0.4 to 200 μm along with the theoretical transmission spectra (dashed black line)[257].

The optical transmission $T(\omega)$ in Fig. 5.2 (b) through N graphene layers on a SiC wafer was expressed in terms of optical conductivity $\sigma(\omega)$ by using the equation below [257]

$$T(\omega) = \left| 1 + N\sigma(\omega)\sqrt{\mu_0/\epsilon_0} / (1 + n_{\text{SiC}}) \right|^{-2}. \quad (5.4)$$

In the far infrared and terahertz spectral range, the optical absorption of graphene mainly stems from intraband transitions. At low frequencies, the conductivity exhibits a Drude-like frequency dependence [260-262]

$$\sigma(\omega) = \frac{\sigma_0}{1 + j\omega\tau}, \quad (5.5)$$

where σ_0 is the low frequency conductivity and τ is the electron relaxation time.

At high frequencies, from mid-infrared to ultraviolet, the absorption is attributed to interband transitions between the valence and conduction bands [263, 264]. The conductivity in this spectral range is not wavelength dependent and within the tight binding model it approaches the value in terms of universal constants given by [265]

$$\sigma(\omega) = \frac{\pi e^2}{2h}. \quad (5.6)$$

This conductivity corresponds to an absorbance of [248, 266]

$$\begin{aligned} A(\omega) &= \frac{4\pi}{c} \sigma(\omega) \\ &= \pi\alpha \\ &\approx 2.3\%, \end{aligned} \quad (5.7)$$

where α is the fine structure constant.

Although the absorption of graphene is nearly constant in this spectral range, it can be controlled by electrostatic gating which shifts the Fermi energy and leads to Pauli blocking [267-270]. In Pauli blocking, the Fermi level is shifted up so that energy levels up to E_F are filled. As a result, optical transitions for pump energies $E_p < 2E_F$ become forbidden as depicted in Fig. 5.3. and graphene becomes transparent for $E_p < 2\Delta E_F$ [66]. By varying the absorption of graphene, it becomes possible to control the loss level introduced into the cavity which becomes important especially inside low-gain laser resonators.

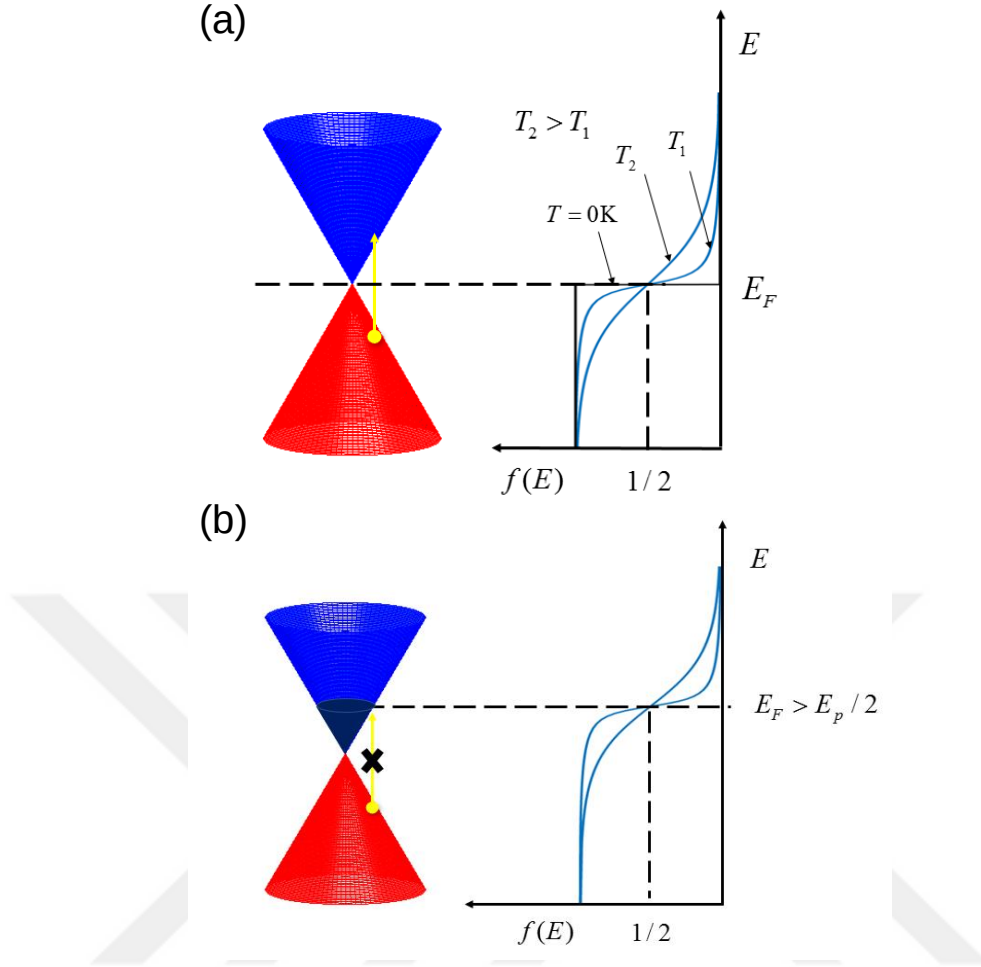


Figure 5.3 Fermi–Dirac distributions of the electrons and the sketch of the energy band diagram for graphene for the case when **(a)** the Fermi level lies at the Dirac point and **(b)** the Fermi level is shifted up.

Next we discuss how the saturation intensity of graphene depends on the photon energy. A simple two-level model can be adopted to describe the saturable absorption in graphene as a function of photocarrier density N [270-272]:

$$\alpha = \frac{\alpha_s}{1 + \frac{N}{N_s}} + \alpha_{NS}, \quad (5.8)$$

where α is the absorption coefficient, α_s is the saturable absorption coefficient, α_{NS} is the nonsaturable absorption, N is the photo-induced carrier density, N_s is the saturation density.

We can express the temporal variation of the photo-carrier density as follows,

$$\frac{\partial N}{\partial t} = \frac{I\alpha}{\hbar\omega} - \frac{N}{\tau} - D\nabla^2 N \quad (5.9)$$

where I is the incident intensity, τ and D are the recombination time and the diffusion coefficient of the carriers, respectively.

The electron-hole recombination out of the excitation region introduced by carrier transfer is built into the last term of Equation 5.9. If the excitation pulsewidth is larger than the carrier recombination time of graphene, the left-hand side of rate Equation 5.9 becomes zero and it reduces to

$$\frac{I\alpha}{\hbar\omega} = \frac{N}{\tau} + D\nabla^2 N. \quad (5.10)$$

Since the experiments in [271] show that the first term on the right side of Equation 5.10 is considerably larger than the second term, we can neglect the carrier diffusion term which reduces the carrier density expression to

$$N = \frac{\alpha I \tau}{\hbar\omega}. \quad (5.11)$$

The substitution of N into Equation 5.8 allows us to express the absorption coefficient in terms of intensity as in the following expression

$$\alpha = \frac{\alpha_s}{1 + \frac{I}{I_s}} + \alpha_{NS}. \quad (5.12)$$

For the transient case where we assume pulse durations (τ_0) are shorter than the carrier recombination time of graphene ($\tau \approx 1.67$ ps), the left-hand side of Eqn 5.9 will become

prominent. If we further assume that the incident beam is Gaussian in spatial and temporal domains, the injected carrier density N will have a Gaussian profile which can be written as [273]

$$N(r, t) = N_0 \exp\left(\frac{-4\ln(2)r^2}{w_0^2}\right) \exp\left(-\frac{t^2}{\tau_0^2}\right) \quad (5.13)$$

where N_0 is the peak carrier density, w_0 and τ_0 are the radius and the pulsewidth of the incident pulse, respectively.

The time derivative of Equation 5.13 can be expressed as follows,

$$\begin{aligned} \frac{\partial N}{\partial t} &= -\frac{2t}{\tau_0^2} N_0 \exp\left(\frac{-4\ln(2)r^2}{w_0^2}\right) \exp\left(-\frac{t^2}{\tau_0^2}\right) \\ &= -\frac{2t}{\tau_0^2} N. \end{aligned} \quad (5.14)$$

Furthermore, the intensity-dependent absorption can be obtained by substitution of equation 5.14 into 5.9

$$\frac{I\alpha}{\hbar\omega} = \frac{2t}{\tau_0^2} N + \frac{N}{\tau} + D\nabla^2 N. \quad (5.15)$$

Alternatively, one can easily see that the saturation intensity for graphene will increase linearly with the photon energy from the well known equation for I_{sa} given by

$$I_{sa} = \frac{h\nu_p}{\sigma_a \tau_f}, \quad (5.16)$$

where I_{sa} is the saturation intensity at the pump wavelength, τ_f is the fluorescence lifetime, σ_a is the stimulated absorption cross section, h is Planck's constant, ν_p is the pump frequency (see Eqn 6.80 in [28]).

Here, note that σ_a and τ_f are nearly constant for graphene hence making I_{sa} proportional to the photon energy $h\nu_p$. From the saturation analysis given above and the experimental results reported in [271], we can conclude that saturable absorption becomes easier for low-energy photons. In other words the saturation intensity decreases as the photon energy decreases. On the other hand, saturable absorption becomes harder for high-energy photons due to high saturation fluence requirement. Therefore, we encounter limited use of graphene saturable absorbers (GSAs) at wavelengths shorter than near-IR spectral region. Graphene as well as graphene-based supercapacitor structures have been utilized as saturable absorbers to generate Q-switched or mode-locked pulses from lasers operating between 800 nm and 3000 nm [269, 274-279]. In particular, graphene was employed in fiber lasers to obtain pulsed output near 3 μm [279]. The shortest mid-infrared pulses were obtained from a graphene mode-locked Cr:ZnS laser at 2.4 μm [280]. Around 1 μm , a graphene based Q-switched fiber laser with 84.1 W average power and 1.67 mJ pulse energy was reported by adopting a master oscillator power amplifier architecture [281]. The repetition rate of 11 GHz was achieved from a cw mode-locked Nd:YAG waveguide laser with a single layer graphene [282]. Near 850 nm, the pulse durations as short as 19 fs were reported from a Cr:LiSAF laser [275]. Baek *et al.* used graphene to mode-lock a Ti:sapphire laser which generated 63 fs pulses at 800 nm [283]. By using a voltage-controlled graphene gold supercapacitor saturable absorber, the mode locking capability of graphene was demonstrated at 795 nm [284].

The use of graphene as a saturable absorber has been scarce at wavelengths below 800 nm and mainly limited to Q-switching [285, 286]. Mode-locking applications of graphene in this spectral range appear more challenging due to the increase of the saturation intensity with decreasing wavelength [271, 276, 283, 287]. Hence, a multi-pass cavity architecture or high-power pumping can be used to increase the intracavity intensities to obtain sufficient absorption saturation for the initiation of mode locking. Several alternative saturable absorbers have been used at wavelengths below 800 nm, including graphene oxide [288], semiconductor saturable absorber mirrors (SESAMs)[289], single-walled carbon nanotubes (SWCNTs)[269], quantum dots (QDs)[207, 290, 291], and emerging two-dimensional (2D) materials[288, 290, 292-315]. Two-dimensional materials such as transition metal-doped dichalcogenides (TMDs), black phosphorus (BPs), and topological insulators (TIs) can provide strong

absorption saturation but may also introduce a high level of nonsaturable loss [314, 316-320], which is undesirable in a low-gain laser medium such as Alexandrite. Furthermore, resonant structures such as SESAMs or QDs typically operate over a relatively narrow spectral window, limiting the operational range of mode-locked lasers.

Recently, there has also been a renewed interest in the development of Alexandrite ($\text{Cr}^{3+}:\text{BeAl}_2\text{O}_4$) lasers, following the emergence of efficient diode or solid-state pump lasers in the visible. To date, efficient pulsed and continuous-wave regimes of operation have been demonstrated [160, 161, 168, 201, 204, 321-323]. Furthermore, by utilizing the broad tuning range of the Alexandrite gain medium, several recent studies have focused on ultrashort pulse generation. In particular, Ghanbari *et al.* reported the generation of femtosecond pulses from a Kerr-lens mode-locked (KLM) Alexandrite laser for the first time [206]. The same group later succeeded in producing 380-fs pulses at 775 nm by using an *InP/InGaP* quantum dot semiconductor saturable absorber mirror [207]. The shortest pulses of 70-fs duration were obtained from a multipass-cavity (MPC) Kerr-lens mode-locked Alexandrite laser [50].

In this chapter, we describe, to the best of our knowledge, the first use of a graphene saturable absorber for the mode locking of an Alexandrite laser. In the experiments, a multi-pass cavity (MPC) extension was added to the short Alexandrite cavity to scale up the pulse energy and also to eliminate the spectral/Q-switching instabilities which were observed in short-length resonators [50, 208]. Pulses with a duration of 65 fs were generated near 750 nm with 5W of pump power. These results further show that graphene can be used as a mode-locker at wavelengths as short as 750 nm.

5.2 Experimental Setup of the graphene mode-locked Alexandrite laser

To demonstrate graphene mode locking of the Alexandrite laser, we adopt a resonator configuration similar to what was previously used for Kerr-lens mode-locking experiments [50]. Figure 5.4 shows a schematic of the experimental setup. Initially, a folded x-cavity was built by using four flat (M3-M6) and two curved (M1 and M2, $R=75$ mm) high reflectors, and a flat output coupler (OC). Then two curved high reflectors (M13-M14, $R=75$ mm) were positioned to create an additional intracavity waist at the location of the graphene saturable absorber (GSA). The monolayer graphene sample used in this work was grown on an infrasil substrate by using chemical vapor deposition. The fast and slow decay

times of the GSA were previously measured to be 128 fs and 1.6 ps, respectively [324]. No active cooling of the GSA was employed in the experiments. The short cavity containing a 10-mm, Brewster-cut, 0.2% Cr-doped Alexandrite crystal (total pump absorption=93% at 532 nm) was end pumped with a single-mode, 5-W, 532-nm pump laser. During the experiments, the crystal was kept inside a copper holder and maintained near 20 °C by water cooling. The end high reflector M6 was later removed and the overall length of the resonator was increased with a notched, multi-pass cavity (MPC). In order to make the MPC q-preserving, the curved (M7, $R=2$ m) and the flat (M8) high reflectors were separated by a distance of 114 cm, and the end high reflector (M12, $R=1$ m) was positioned at a distance of 114 cm from M8. The design rules of this class of notched, q-preserving MPCs was discussed in earlier studies [85]. With the MPC, an additional optical path length of 50.2 m was added to the Alexandrite resonator, giving a pulse repetition frequency of 5.56 MHz.

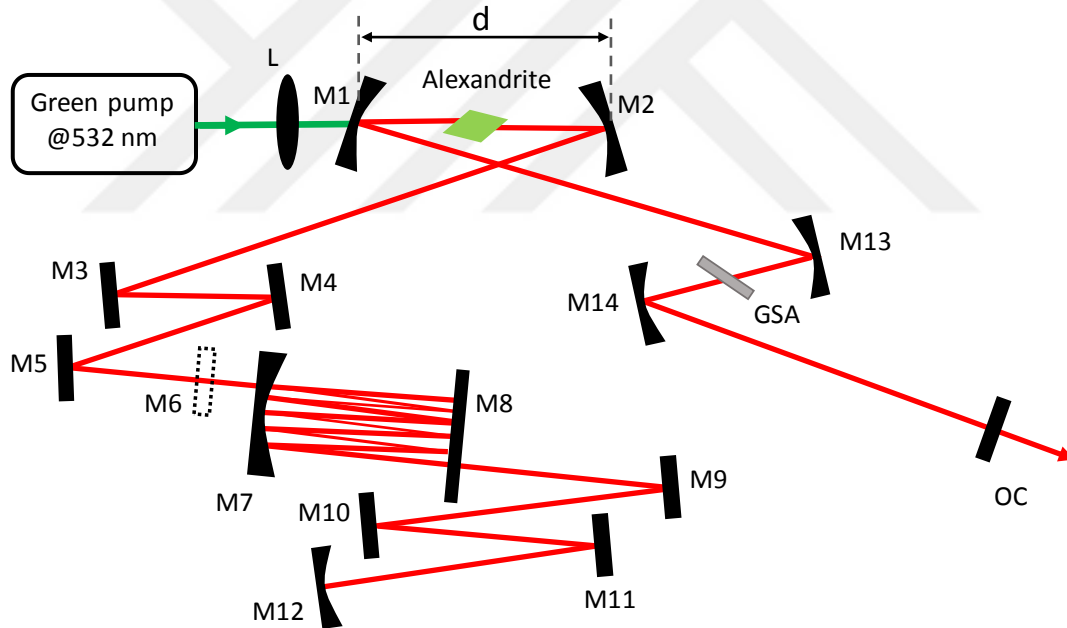


Figure 5. 4 Schematic of the 532-nm pumped, multipass-cavity femtosecond Alexandrite laser, mode-locked near 750 nm with a monolayer graphene saturable absorber.

5.3 Continuous-wave Operation Results

The continuous-wave power performance of the Alexandrite laser was investigated for the short and extended resonators. Figure 5.5(a) shows the measured power efficiency of the x-cavity Alexandrite laser by using four output couplers with 0.1%, 0.25%, 0.5%, and 1.7% transmission near 750 nm. The resulting measured slope efficiencies with four different output couplers were 2.8%, 5.9%, 11.4%, and 13.9%, respectively. The roundtrip passive loss of the x-cavity Alexandrite laser was estimated to be 0.61% by using the threshold pump power data shown in Fig. 5.5(b).

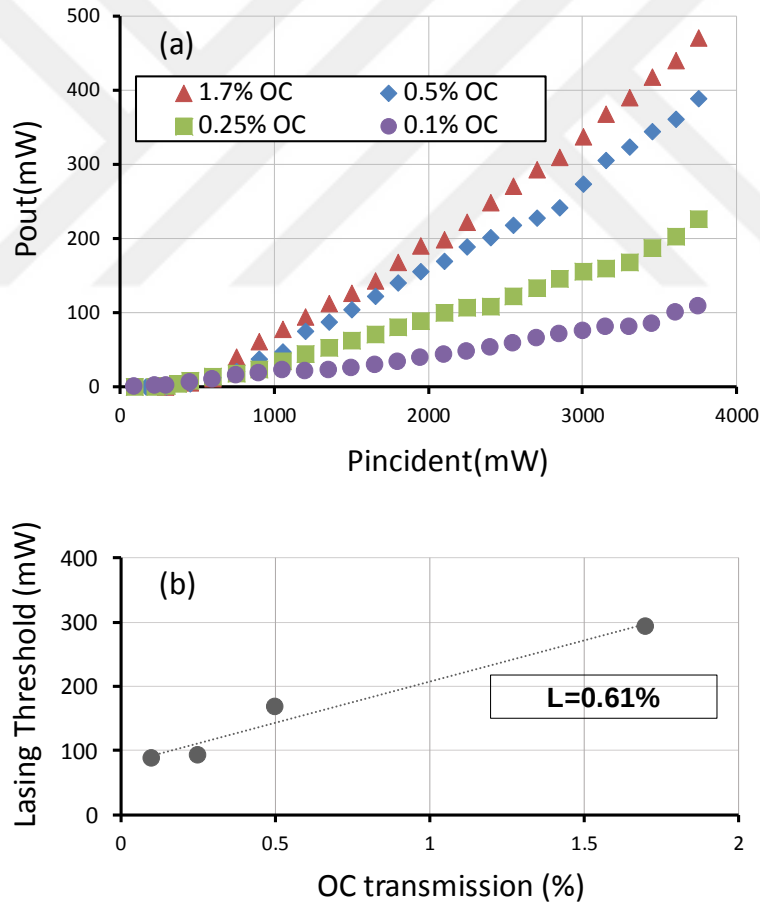


Figure 5. 5 (a) Continuous-wave power efficiency curves of the x-cavity Alexandrite laser measured with four output couplers. **(b)** Measured threshold pump power as a function of output coupler transmission.

Figure 5.6 shows the continuous-wave power efficiency data of the short-cavity and extended-cavity Alexandrite laser operating with the 0.25% output coupler. The MPC extension decreased the power slope efficiency of the x-cavity Alexandrite laser from 5.9% to 1.7%. The insertion of the graphene sample further decreased the slope efficiency of the laser to 0.7%. The roundtrip passive loss introduced by the MPC and the graphene saturable absorber was estimated to be 3.05% and 4.82%, respectively, based on the threshold data in Figure 5.6.

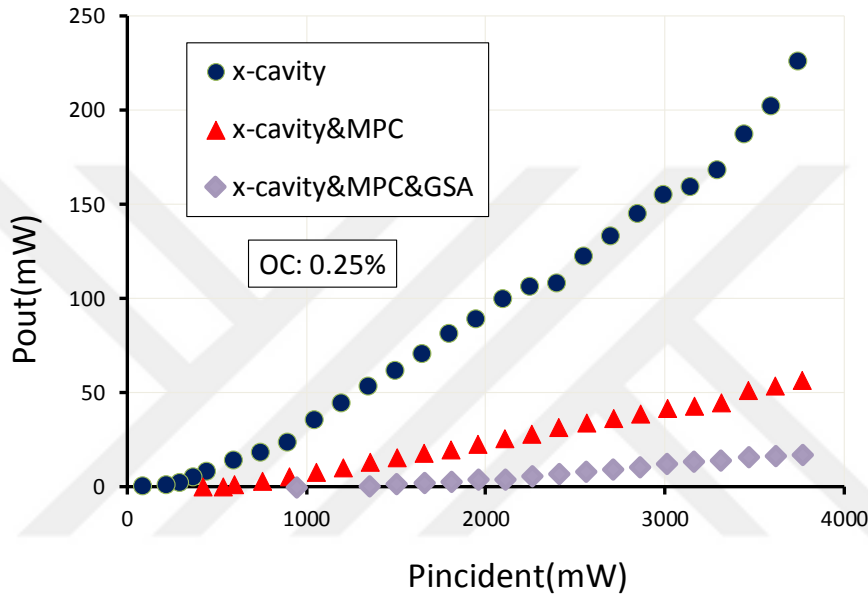


Figure 5.6 The measured power efficiency of the short-, multipass-cavity, and graphene saturable absorber added multipass-cavity Alexandrite laser operating with the 0.25% output coupler.

5.4 Mode-locked Operation Results

In the mode-locking experiments described here, it is important to ensure that pulse generation is indeed initiated via the saturable absorber action in graphene and not by Kerr-lens mode locking (KLM). To investigate the cavity configurations for graphene mode locking and KLM, the separation (d in Fig. 5.4) between the curved mirrors M1 and M2 was varied at 10- μm steps and the output power of the laser was measured across the full stability range of the resonator. At each M1-M2 separation, the output coupler was translated to see whether mode locking could be initiated. Two separate cases were investigated. In the first case, GSA transferred onto an infrasil substrate was located at the waist position between M13 and M14, and the output power was measured as a function of d at the input pump power of 5W (Fig. 5.7(a)). The separations where mode locking could be initiated are marked with solid red

triangles in Fig. 5.7(a). Note that when GSA was used, mode locking could be initiated nearly across the full stability range. In the second case, a bare infrasil substrate without graphene was placed between M13 and M14. To keep the output power and, hence, the intracavity pulse energy at about the same level as in the first case, the input pump power was reduced to 2.7 W. Here, the mode-locking initiation mechanism is Kerr lensing. Note that KLM action could be observed at fewer points due to the requirement for critical cavity alignment to initiate Kerr lensing (Fig. 5.7(b)). Based on the data shown in Figs. 5.7(a) and (b), graphene mode-locking data of the Alexandrite laser were recorded near $d=8.161$ cm, where KLM action was not observed with bare infrasil substrate. In the mode-locking experiments, negative group delay dispersion (GDD) was provided by most of the cavity mirrors, including the MPC mirrors. The dispersion control mirrors M4, M5, M7, M8, M9, and M12 each had a GDD of -50 fs^2 per bounce, whereas M10 and M11 had a GDD of -100 fs^2 , and M3 had a GDD of -40 fs^2 per bounce. By taking into account the GDD due to the gain medium (60 fs^2 per mm [75]), air (20 fs^2 per meter), the infrasil substrate ($\sim 100 \text{ fs}^2$ per 1 mm), the mirrors M1, M2, and OC ($\sim 5 \text{ fs}^2$ per bounce), as well as the mirrors M13- M14 ($\sim 25 \text{ fs}^2$ per bounce), the net round-trip GDD of the composite resonator shown in Fig. 5.4 was estimated to be around -400 fs^2 near 750 nm.

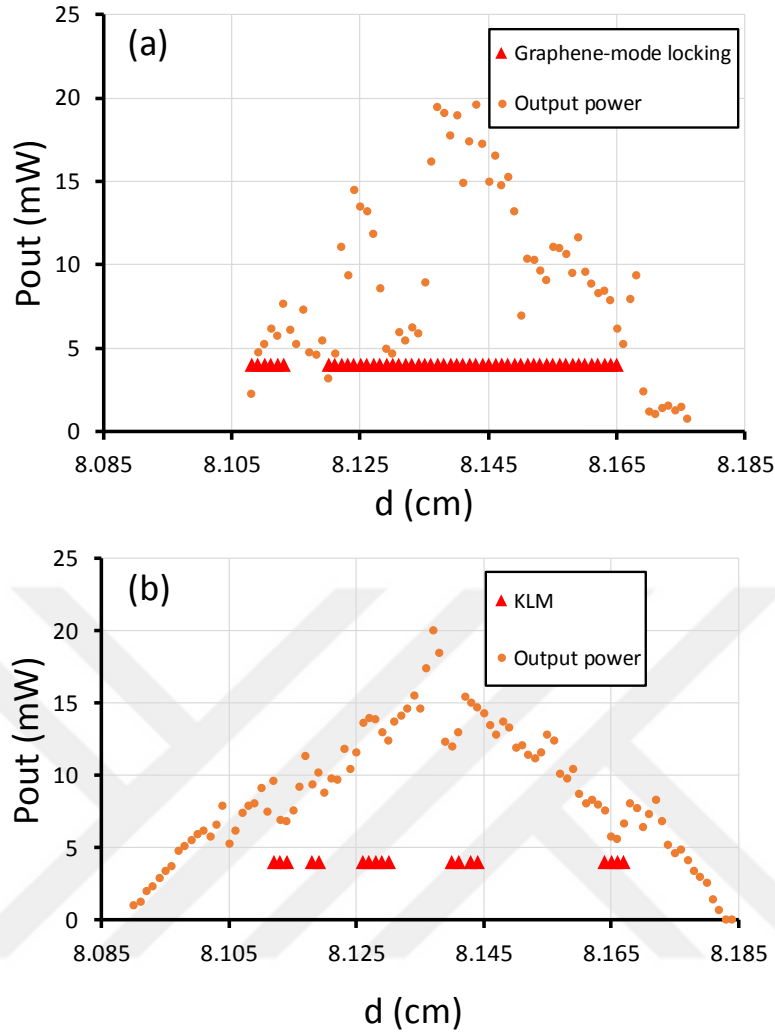


Figure 5. 7 Measured output power and mode-locking tendency (solid triangles) of the Alexandrite laser as a function of the M2-M3 separation position for **(a)** GSA and **(b)** bare infrasil substrate between M13 and M14

Figures 5.8 and 5.9 show the mode-locking results obtained from the MPC Alexandrite laser. Figure 5.8(a) shows the interferometric autocorrelation trace of the graphene mode-locked Alexandrite laser. The temporal duration of the pulses was determined to be 65 fs, by assuming a sech^2 intensity profile. Figure 5.8(b) further shows the measured optical spectrum of the generated pulses and the estimated round-trip GDD of the resonator as a function of wavelength. The spectral width (FWHM) was measured to be 9.3 nm. The measured time-bandwidth product of 0.319 suggests that the pulses were nearly transform limited. Note that the net group delay dispersion of the resonator was positive at wavelengths below 740 nm, preventing solitary pulse generation with spectra that span the full tuning range of the Alexandrite laser. We foresee that by using cavity optics with optimized GDD over the entire tuning range, it should be possible to generate sub-10 fs pulses from the Alexandrite laser [161].

At the pump power of 5W, the output power of the laser during mode-locked operation was 8 mW, corresponding to 1.4 nJ of pulse energy at the pulse repetition frequency of 5.56 MHz. The peak power was 22 kW. Based on the ABCD analysis of the cavity, the spotsizes on the GSA was estimated to be 25 mm, yielding an incident fluence of 20 mJ/cm^2 on the GSA. Figures 5.9(a) and 5.9(b) further show the measured radio-frequency (RF) spectrum and the pulse train of the graphene mode-locked MPC Alexandrite laser. As can be seen from Fig. 5.9(a), the pulse train carrier signal at 5.56 MHz had a 75-dBc contrast with respect to the noise background, indicating mode-locked operation without Q-switching instabilities.

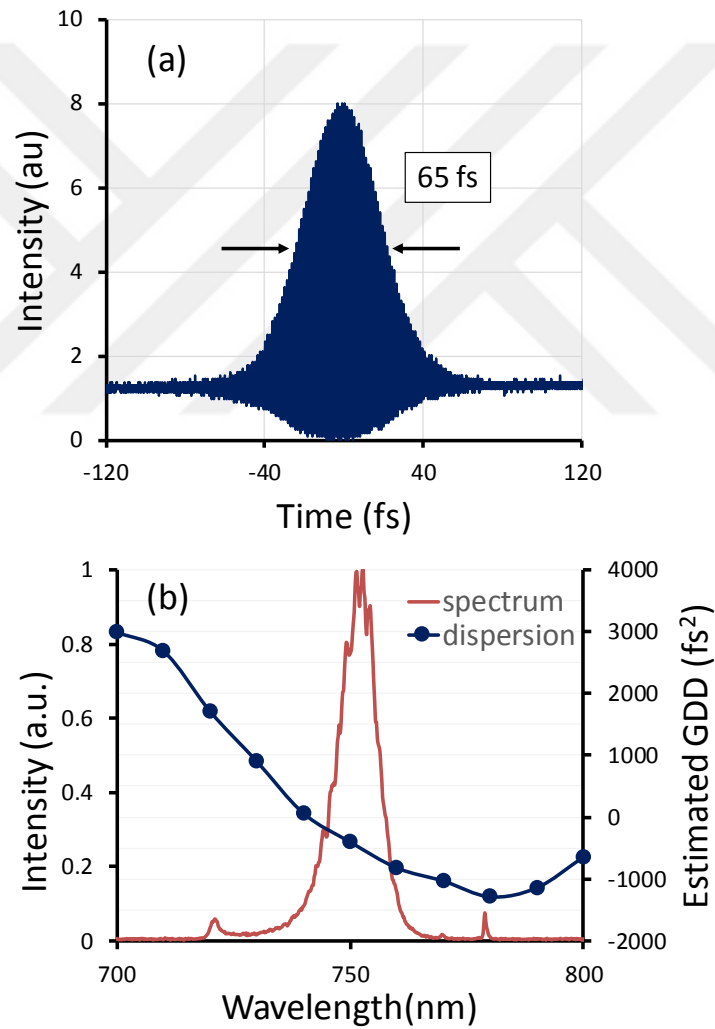


Figure 5. 8 (a) The interferometric autocorrelation trace of the 65-fs pulses obtained from the graphene mode-locked Alexandrite laser. **(b)** The measured optical spectrum and estimated group delay dispersion (GDD)

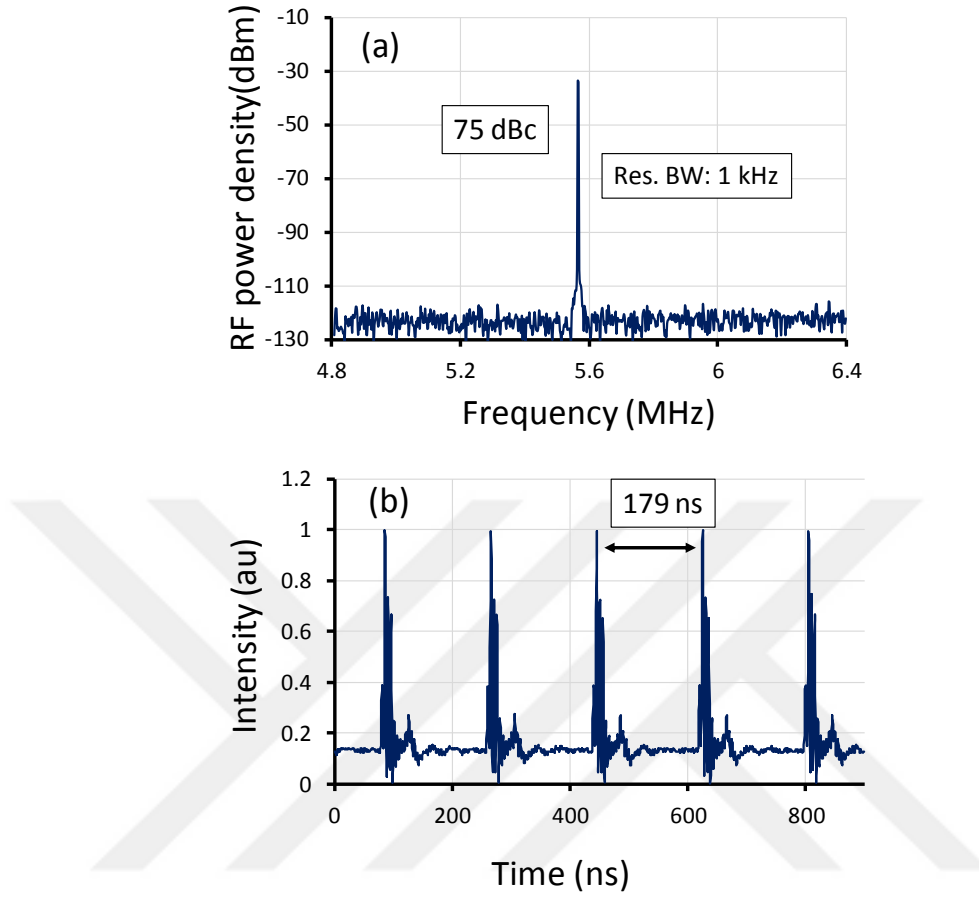


Figure 5. 9 The measured **(a)** radio frequency (RF) spectrum and **(b)** pulse train of the pulses generated with the graphene mode-locked Alexandrite laser at the repetition rate of 5.56 MHz. During the RF spectrum measurements, the resolution bandwidth (Res. BW) was 1kHz.

In conclusion, we report, for the first time to our knowledge, graphene mode-locked operation of a femtosecond Alexandrite laser. By using a multipass cavity architecture and with 5W of pump power at 532 nm, we obtained 65-fs, nearly transform-limited pulses at 750 nm. The RF peak at the repetition frequency of 5.56 MHz had a contrast of 75 dBc with respect to the background noise level. The energy, peak power, and the time-bandwidth product of the generated pulses were 1.4 nJ, 22 kW, and 0.319 respectively. Finally, this represents the shortest wavelength, to our knowledge, where graphene has been utilized to initiate mode-locked operation of a laser.

Chapter 6

Conclusions

In this dissertation, we presented our femtosecond pulse generation results obtained from Cr:LiSAF and Alexandrite lasers. Fundamental spectroscopic and laser characteristics of both gain media were compared with that of Ti:sapphire. Despite the fact that the nonlinear refractive index of both gain media was lower than that of Ti:sapphire, we could successfully generate femtosecond pulses from Cr:LiSAF and Alexandrite gain media. For the Kerr-lens mode locking of tapered-diode pumped (1W, 680 nm) Cr:LiSAF laser, we used a gain matched output coupler (GMOC) which resulted in an efficient and robust KLM operation around 850 nm. For the Kerr-lens mode locking of green pumped (1W, 532 nm) Alexandrite laser, we used a multipass cavity extension to eliminate spectral instabilities observed in the short cavity. This enabled us to obtain the shortest femtosecond pulses generated from an Alexandrite laser reported to date. Furthermore, we adopted a similar resonator configuration and utilised graphene as a saturable absorber to generate sub-100-fs pulses from green pumped (5W, 532 nm) Alexandrite laser near 750 nm. This was the shortest femtosecond pulse generation wavelength with graphene saturable absorber reported to date.

In Chapter 2, we reviewed some fundamental theoretical concepts such as passive mode locking methods (Kerr-lens and saturable absorber mode locking), group velocity dispersion, and soliton area theorem. This was followed by the discussion about population inversion and lasing conditions in a four-level atomic system. We ended the chapter with the design principles of q-preserving multipass cavities. This chapter established a brief background for the experimental studies performed in this thesis.

In Chapter 3, we demonstrated efficient and robust Kerr-lens mode-locking of tapered diode-pumped Cr:LiSAF laser by using a gain-matched output coupler (GMOC). The Tapered diode (TD) laser which was fabricated and characterized at the facilities of the Ferdinand Braun Institute was used as a pump source. TD could provide up to 1050 mW of output power at 680

nm with an electrical-to-optical conversion efficiency of 22 %. We introduced a specially designed output coupler GMOC into the Cr:LiSAF cavity. The transmission profile of the GMOC matched the gain spectrum of the laser active medium. This generated a loss profile that had the same shape with the cavity gain which resulted in a flat net gain profile. As a result, the need for strong SAM was eliminated and a low level of KLM action became sufficient to sustain ultrashort pulses. In our experiments, with the use of GMOC technology, long-term stable and robust KLM operation could be achieved. Once the KLM was initiated by slightly perturbing the output coupler, the laser remained in mode-locked operation for hours. The average output power of 106 mW was obtained at an incident pump power of 940 mW. TD pumped Cr:LiSAF laser generated sub-15-fs pulses with peak powers of 60-kW at repetition rate of 113 MHz. This corresponded to pulse energies and peak powers of 940 pJ and 60 kW, respectively. Further, a fs tuning range from 807nm to 919 nm was demonstrated with sub-50-fs pulses. The obtained results in this study clearly demonstrate the usefulness of the GMOC mirrors in reducing the stringent requirements of Kerr lens mode locking even in gain media with a low nonlinearity such as Cr:LiSAF.

In Chapter 4, we focused on the generation of the shortest pulses obtained from a Kerr-lens mode-locked multipass-cavity Alexandrite laser operating near 750 nm. This was achieved by using an extended optical resonator. The increase in the intracavity pulse energies eliminated the spectral/Q-switching instabilities and enabled cw mode-locked operation. We also emphasized the design principles of the q-preserving multipass-cavity extension. Continuous wave, 1W green laser at 532 nm was used to pump multipass-cavity Alexandrite laser. Both short cavity and extended cavity power performances were analysed. During Kerr-lens mode-locked operation, nearly transform-limited 70-fs pulses could be obtained with 0.7 nJ of energy per pulse. As was mentioned in the chapter, the net group delay dispersion of the resonator was positive at wavelengths below 740 nm. This prevented solitary pulse generation with spectra that span the full tuning range of the laser. Hence, we expect that further dispersion optimization of the resonator could result in shorter pulses. These results suggest that Alexandrite lasers have the potential of becoming versatile sources in the near-infrared spectral range.

In Chapter 5, we described the femtosecond pulse generation from a graphene mode-locked Alexandrite laser for the first time. The extended-cavity Alexandrite laser produced 65-fs nearly transform-limited pulses with 5W of green pump power at 532 nm. The RF peak at the repetition frequency of 5.56 MHz had a contrast of 75 dBc with respect to the noise floor. The energy, peak power, and the time-bandwidth product of the generated pulses were 1.4 nJ,

22 kW, and 0.319 respectively. Graphene with its ultrabroad operation bandwidth and ultrashort response time was utilised in a wide range of lasers. However, the increase of the saturation intensity of graphene in shorter wavelengths hampered its use in the visible. In our study, we demonstrated the operation of graphene as a saturable absorber/mode-locker at wavelength as short as 750 nm which is the lowest wavelength reported to date.

We believe that the experimental studies demonstrated in this dissertation will contribute to the development of low-cost, efficient, and robust tunable femtosecond laser sources for various potential applications in the near-infrared spectral region. In this regard, Alexandrite and Cr:LiSAF lasers can be utilised as good alternatives to Ti:sapphire around 800 nm with the help of the techniques described in the experimental parts of this thesis.



Appendix

Interferometric Autocorrelation

In this section, the operation of an autocorrelator for pulsewidth characterization is described. Interferometric autocorrelation is a widely used technique to measure the duration of ultrashort pulses [325-329]. Figure A.1 shows the autocorrelator setup that we used to measure the pulse durations. The setup consists of a Michelson interferometer, a non-linear crystal for second harmonic generation and a detector. The incoming pulse is split into two replicas and one of them is given a time delay. Then, they are recombined with a second beam splitter and passed through a second harmonic generation crystal. After the crystal, the intensity of the generated second harmonic signal is detected with a detector. The resulting intensity of the second harmonic signal is given by the superposition of two identical pulses delayed by τ with respect to each other

$$E(t, \tau) = E(t + \tau) + E(t). \quad (\text{A.1})$$

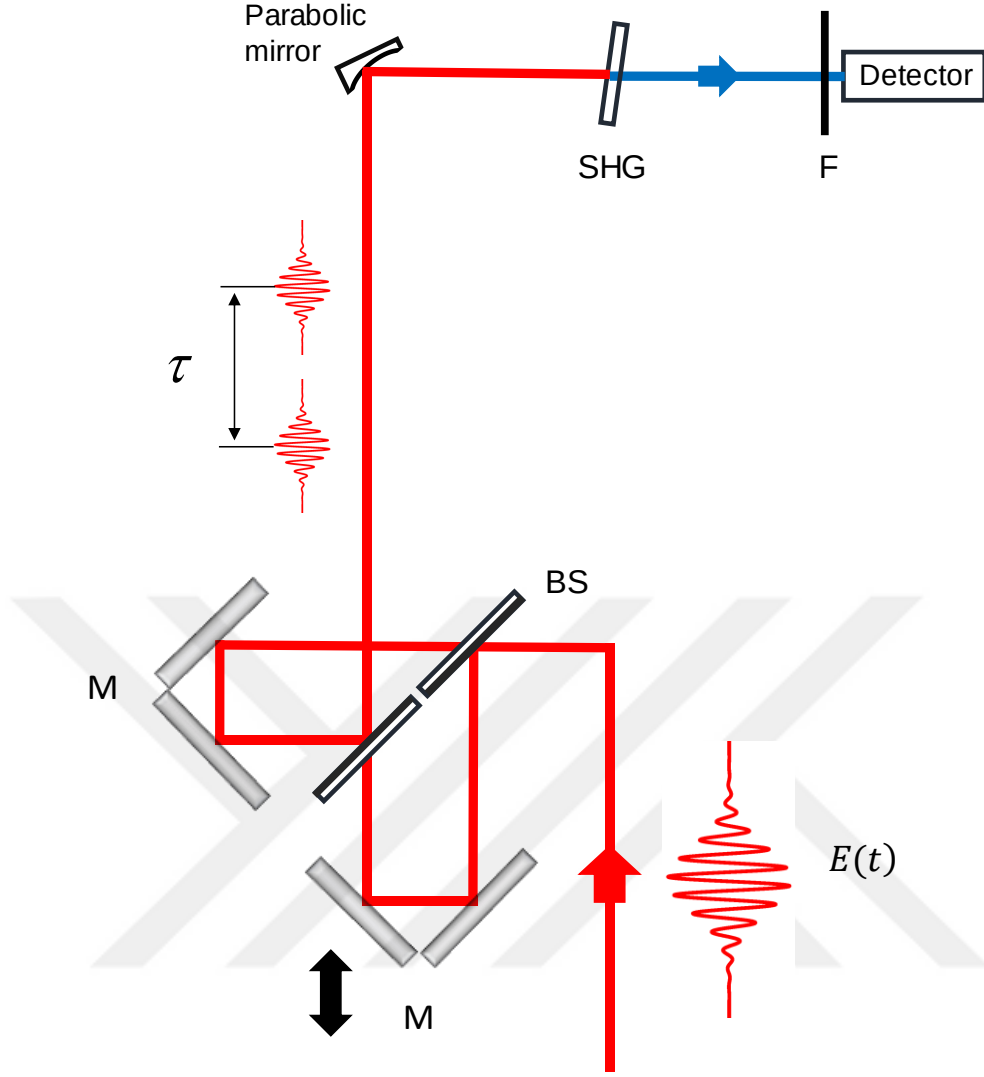


Figure A. 1 Optical layout for the autocorrelation setup.

The second harmonic electric field is proportional to the square of the total field at the fundamental frequency. The signal intensity detected by the photo detector is given by [76]

$$I(\tau) \propto \int_{-\infty}^{\infty} \left| \left(A(t+\tau)e^{j\omega_c(t+\tau)} + A(t)e^{j\omega_c t} \right)^2 \right|^2 dt, \quad (\text{A.2})$$

$$I(\tau) \propto \int_{-\infty}^{\infty} \left| A^2(t+\tau)e^{j2\omega_c(t+\tau)} + 2A(t+\tau)A(t)e^{j\omega_c(t+\tau)}e^{j\omega_c t} + A^2(t)e^{j2\omega_c t} \right|^2 dt.$$

We can expand the expression given above as in Eqn. A.3 with the help of the identity $(|z| = (z^* z)^{1/2})$ [76]:

$$\begin{aligned}
 I(\tau) \propto & \int_{-\infty}^{+\infty} [|A(t+\tau)|^4 + 4|A(t+\tau)|^2 |A(t)|^2 + |A^2(t)|^2 \\
 & + 2|A(t+\tau)|^2 A(t+\tau)A^*(t)e^{j\omega_c\tau} + 2|A(t+\tau)|^2 A^*(t+\tau)A(t)e^{-j\omega_c\tau} \\
 & + 2|A(t)|^2 A(t+\tau)A^*(t)e^{j\omega_c\tau} + 2|A(t)|^2 A^*(t+\tau)A(t)e^{-j\omega_c\tau} \\
 & + A^2(t+\tau)(A^*(t))^2 e^{j2\omega_c\tau} + A^2(t)(A^*(t+\tau))^2 e^{-j2\omega_c\tau}] dt.
 \end{aligned} \tag{A.3}$$

The interferometric autocorrelation function above is normalized relative to the background intensity given by

$$I_{back} = \int_{-\infty}^{\infty} (|A(t+\tau)|^4 + |A(t)|^4) dt. \tag{A.4}$$

For $\tau = 0$ and $\tau \rightarrow \pm\infty$, we obtain the ratio of 8:1 between the maxima and the wings of the interferometric autocorrelation function

$$\begin{aligned}
 I_{AC}(0) &= 8 \\
 I_{AC}(\tau \rightarrow \pm\infty) &= 1.
 \end{aligned} \tag{A.5}$$

The proper alignment of the autocorrelator can be checked with the obtained center-to-background ratios. The interferometric autocorrelation does not give sufficient information to characterize arbitrary-shaped and few cycle ultrashort laser pulses. More sophisticated techniques such as SPIDER and FROG have emerged for the complete characterization of the pulse shape [327, 330, 331].

Bibliography

1. A. Einstein, "Zur Quantentheorie der Strahlung," Physikalische Gesellschaft Zürich. Mitteilungen (1916).
2. J. P. Gordon, H. J. Zeiger, and C. H. Townes, "The Maser---New Type of Microwave Amplifier, Frequency Standard, and Spectrometer," *Physical Review* **99**, 1264-1274 (1955).
3. A. Javan, W. R. Bennett, and D. R. Herriott, "Population Inversion and Continuous Optical Maser Oscillation in a Gas Discharge Containing a He-Ne Mixture," *Physical Review Letters* **6**, 106-110 (1961).
4. M. G. A. Bernard and G. Durauffourg, "Laser Conditions in Semiconductors," *physica status solidi (b)* **1**, 699-703 (1961).
5. J. E. Geusic, "Laser Oscillations in Nd-Doped Yttrium Aluminum, Yttrium Gallium and Gadolinium Garnets," *Applied Physics Letters* **4**, 182-184 (1964).
6. L. D. Smullin and G. Fiocco, "Optical Echoes from the Moon," *Nature* **194**, 1267 (1962).
7. G. Neumann, "National Aeronautics and Space Administration Goddard Space Flight Center", retrieved 2018, <https://tharsis.gsfc.nasa.gov/MOLA/mola.php>.
8. C. Weitkamp, *Lidar: Range-Resolved Optical Remote Sensing of the Atmosphere* (Springer New York, 2006).
9. M. Hentschel, R. Kienberger, C. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, "Attosecond metrology," *Nature* **414**, 509 (2001).
10. A. J. DeMaria, "Mode Locking of a Nd³⁺-Doped Glass Laser," *Applied Physics Letters* **8**, 22-24 (1966).
11. J. Valdmanis and R. Fork, "Design considerations for a femtosecond pulse laser balancing self phase modulation, group velocity dispersion, saturable absorption, and saturable gain," *IEEE Journal of Quantum Electronics* **22**, 112-118 (1986).
12. D. E. Spence, Kean, P. N., Sibbett, W., "60-fsec pulse generation from a self-mode-locked Ti:sapphire laser," *Opt. Lett.* **16**(1991).
13. F. Krausz, M. E. Fermann, T. Brabec, P. F. Curley, M. Hofer, M. H. Ober, C. Spielmann, E. Wintner, and A. J. Schmidt, "Femtosecond solid-state lasers," *IEEE Journal of Quantum Electronics* **28**, 2097-2122 (1992).
14. L. E. Hargrove, "Locking of He-Ne Laser Modes Induced By Synchronous Intracavity Modulation," *Applied Physics Letters* **5**, 4-5 (1964).
15. M. D. Jr, "Small-Signal Analysis of Internal (Coupling-Type) Modulation of Lasers," *Journal of Applied Physics* **35**, 2870-2876 (1964).
16. R. L. Fork, "Generation of optical pulses shorter than 0.1 psec by colliding pulse mode locking," *Applied Physics Letters* **38**, 671-672 (1981).
17. W. Denk, J. H. Strickler, and W. W. Webb, "Two-Photon Laser Scanning Fluorescence Microscopy," *Science* **248**, 73-76 (1990).
18. B. E. A. Saleh and M. C. Teich, *Fundamentals of Photonics* (Wiley, 2013).
19. S. Ruzin and H. Aaron, "https://neuropsychics.ucsd.edu/courses/physics_173_273/2014_FinalReport_NonlinearOptics.pdf", retrieved 2018.

Bibliography

20. A. Diaspro, P. Bianchini, G. Vicidomini, M. Faretta, P. Ramoino, and C. Usai, "Multi-photon excitation microscopy," *BioMedical Engineering OnLine* **5**, 36 (2006).
21. S. Sakadžić, U. Demirbas, T. R. Mempel, A. Moore, S. Ruvinskaya, D. A. Boas, A. Sennaroglu, F. X. Kartner, and J. G. Fujimoto, "Multi-photon microscopy with a low-cost and highly efficient Cr:LiCAF laser," *Opt. Express* **16**, 20848-20863 (2008).
22. C. Xu and W. W. Webb, "Measurement of two-photon excitation cross sections of molecular fluorophores with data from 690 to 1050 nm," *J. Opt. Soc. Am. B* **13**, 481-491 (1996).
23. W. Denk, D. W. Piston, and W. W. Webb, "Two-Photon Molecular Excitation in Laser-Scanning Microscopy," in *Handbook of Biological Confocal Microscopy*, J. B. Pawley, ed. (Springer US, Boston, MA, 1995), pp. 445-458.
24. P. Xi, Y. Andegeko, L. R. Weisel, V. V. Lozovoy, and M. Dantus, "Greater signal, increased depth, and less photobleaching in two-photon microscopy with 10fs pulses," *Optics Communications* **281**, 1841-1849 (2008).
25. P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich, "Generation of Optical Harmonics," *Physical Review Letters* **7**, 118-119 (1961).
26. N. Bloembergen and Y. R. Shen, "Quantum-Theoretical Comparison of Nonlinear Susceptibilities in Parametric Media, Lasers, and Raman Lasers," *Physical Review* **133**, A37-A49 (1964).
27. J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, "Interactions between Light Waves in a Nonlinear Dielectric," *Physical Review* **127**, 1918-1939 (1962).
28. A. Sennaroglu, *Photonics and laser engineering : principles, devices, and applications* (McGraw-Hill, New York, 2010).
29. A. Yariv and P. Yeh, *Photonics: Optical Electronics in Modern Communications* (Oxford University Press, Incorporated, 2007).
30. R. W. Boyd, "Chapter 1 - The Nonlinear Optical Susceptibility," in *Nonlinear Optics (Second Edition)*, R. W. Boyd, ed. (Academic Press, San Diego, 2003), pp. 1-65.
31. C. Jin, G. Wang, A.-T. Le, and C. D. Lin, "Route to optimal generation of soft X-ray high harmonics with synthesized two-color laser pulses," *Scientific Reports* **4**, 7067 (2014).
32. T. Popmintchev, M.-C. Chen, D. Popmintchev, P. Arpin, S. Brown, S. Ališauskas, G. Andriukaitis, T. Balčiunas, O. D. Mücke, A. Pugzlys, A. Baltuška, B. Shim, S. E. Schrauth, A. Gaeta, C. Hernández-García, L. Plaja, A. Becker, A. Jaron-Becker, M. M. Murnane, and H. C. Kapteyn, "Bright Coherent Ultrahigh Harmonics in the keV X-ray Regime from Mid-Infrared Femtosecond Lasers," *Science* **336**, 1287-1291 (2012).
33. T. H. Maiman, "Stimulated Optical Radiation in Ruby," *Nature* **187**, 493 (1960).
34. W. R. Zipfel, R. M. Williams, and W. W. Webb, "Nonlinear magic: multiphoton microscopy in the biosciences," *Nature Biotechnology* **21**, 1369 (2003).
35. A. Bartels, T. Dekorsy, and H. Kurz, "Femtosecond Ti:sapphire ring laser with a 2-GHz repetition rate and its application in time-resolved spectroscopy," *Opt. Lett.* **24**, 996-998 (1999).
36. T. Udem, J. Reichert, R. Holzwarth, and T. W. Hänsch, "Absolute Optical Frequency Measurement of the Cesium D₁ Line with a Mode-Locked Laser," *Physical Review Letters* **82**, 3568-3571 (1999).
37. T. Udem, R. Holzwarth, and T. W. Hänsch, "Optical frequency metrology," *Nature* **416**, 233 (2002).
38. L. F. Johnson, R. E. Dietz, and H. J. Guggenheim, "Optical maser oscillation from Ni²⁺ in MgF₂ involving simultaneous emission of phonons," *Physical Review Letters* **11**, 318-320 (1963).
39. L. F. Mollenauer and D. H. Olson, "Broadly tunable lasers using color centers," *Journal of Applied Physics* **46**, 3109-3118 (1975).
40. J. C. Walling, H. P. Jenssen, R. C. Morris, E. W. O'Dell, and O. G. Peterson, "Tunable-laser performance in BeAl₂O₄:Cr³⁺," *Opt. Lett.* **4**, 182-183 (1979).
41. P. Moulton, "Ti-doped sapphire: tunable solid-state laser," *Optics News* **8**, 9-9 (1982).
42. J. Harrison, A. Finch, D. M. Rines, G. A. Rines, and P. F. Moulton, "Low-threshold, cw, all-solid-state Ti:Al₂O₃ laser," *Opt. Lett.* **16**, 581-583 (1991).

43. R. Ell, U. Morgner, F. X. Kärtner, J. G. Fujimoto, E. P. Ippen, V. Scheuer, G. Angelow, T. Tschudi, M. J. Lederer, A. Boiko, and B. Luther-Davies, "Generation of 5-fs pulses and octave-spanning spectra directly from a Ti:sapphire laser," *Opt. Lett.* **26**, 373-375 (2001).
44. P. F. Moulton, "Spectroscopic and laser characteristics of Ti:Al₂O₃," *J. Opt. Soc. Am. B* **3**, 125-133 (1986).
45. F. Druon, F. Balembois, and P. Georges, "New laser crystals for the generation of ultrashort pulses," *Comptes Rendus Physique* **8**, 153-164 (2007).
46. W. R. Rapoport and C. P. Khatkhaty, "Titanium sapphire laser characteristics," *Appl. Opt.* **27**, 2677-2684 (1988).
47. W. Koechner, *Solid-State Laser Engineering* (Springer New York, 2007).
48. S. A. Payne, L. L. Chase, L. K. Smith, W. L. Kway, and H. W. Newkirk, "Laser performance of LiSrAlF₆:Cr³⁺," *Journal of Applied Physics* **66**, 1051-1056 (1989).
49. C. Cihan, E. Beyatli, F. Canbaz, L. J. Chen, B. Sumpf, G. Erbert, A. Leitenstorfer, F. X. Kärtner, A. Sennaroglu, and U. Demirbas, "Gain-Matched Output Couplers for Efficient Kerr-Lens Mode-Locking of Low-Cost and High-Peak Power Cr:LiSAF Lasers," *IEEE Journal of Selected Topics in Quantum Electronics* **21**(2015).
50. C. Cihan, A. Muti, I. Baylam, A. Kocabas, U. Demirbas, and A. Sennaroglu, "70 femtosecond Kerr-lens mode-locked multipass-cavity Alexandrite laser," *Opt. Lett.* **43**, 1315-1318 (2018).
51. C. Cihan, C. Kocabas, U. Demirbas, and A. Sennaroglu, "Graphene mode-locked femtosecond Alexandrite laser," *Opt. Lett.* **43**, 3969-3972 (2018).
52. P. B. Corkum and F. Krausz, "Attosecond science," *Nature Physics* **3**, 381 (2007).
53. L. E. Hargrove, R. L. Fork, and M. A. Pollack, "Locking of He-Ne Laser Modes Induced By Synchronous Intracavity Modulation," *Applied Physics Letters* **5**, 4-5 (1964).
54. M. Hentschel, R. Kienberger, S. Ch. G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, "Attosecond metrology," *Nature* **414**, 509-513 (2001).
55. P. M. W. French, "The generation of ultrashort laser pulses," *Reports on Progress in Physics* **58**, 169 (1995).
56. G. H. C. New, "The generation of ultrashort laser pulses," *Reports on Progress in Physics* **46**, 877 (1983).
57. J. D. Simon, "Ultrashort light pulses," *Review of Scientific Instruments* **60**, 3597-3624 (1989).
58. H. A. Haus, "Mode-locking of lasers," *IEEE Journal of Selected Topics in Quantum Electronics* **6**, 1173-1185 (2000).
59. K. Gurs, "Modulation and mode locking of the continuous ruby laser," *IEEE Journal of Quantum Electronics* **3**, 175-180 (1967).
60. J. Kerr, "LIV. A new relation between electricity and light: Dielectrified media birefringent (Second paper)," *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science* **50**, 446-458 (1875).
61. D. E. Spence, P. N. Kean, and W. Sibbett, "60-fsec pulse generation from a self-mode-locked Ti:sapphire laser," *Opt. Lett.* **16**, 42-44 (1991).
62. L. J. Qian, X. Liu, and F. W. Wise, "Femtosecond Kerr-lens mode locking with negative nonlinear phase shifts," *Opt. Lett.* **24**, 166-168 (1999).
63. H. A. Haus, J. G. Fujimoto, and E. P. Ippen, "Analytic theory of additive pulse and Kerr lens mode locking," *IEEE Journal of Quantum Electronics* **28**, 2086-2096 (1992).
64. A. Sennaroglu, C. R. Pollock, and H. Nathel, "Continuous-wave self-mode-locked operation of a femtosecond Cr⁴⁺:YAG laser," *Opt. Lett.* **19**, 390-392 (1994).
65. A. Seas, V. Petričević, and R. R. Alfano, "Self-mode-locked chromium-doped forsterite laser generates 50-fs pulses," *Opt. Lett.* **18**, 891-893 (1993).
66. I. Baylam, S. Özharar, N. Kakenov, C. Kocabaş, and A. Sennaroglu, "Femtosecond Pulse Generation with Voltage-Controlled Graphene Saturable Absorbers," in *Optical Properties of Graphene*, pp. 389-433.
67. F. Träger, *Springer Handbook of Lasers and Optics* (Springer Berlin Heidelberg, 2012).

68. H. A. Haus, "Theory of mode locking with a fast saturable absorber," *Journal of Applied Physics* **46**, 3049-3058 (1975).
69. H. Haus, "Theory of mode locking with a slow saturable absorber," *IEEE Journal of Quantum Electronics* **11**, 736-746 (1975).
70. A. Sennaroglu, *Solid-State Lasers and Applications* (Taylor & Francis, 2010).
71. J. C. Diels, W. Rudolph, P. F. Liao, and P. Kelley, *Ultrashort Laser Pulse Phenomena: Fundamentals, Techniques, and Applications on a Femtosecond Time Scale* (Elsevier Science, 2006).
72. R. L. Fork, O. E. Martinez, and J. P. Gordon, "Negative dispersion using pairs of prisms," *Opt. Lett.* **9**, 150-152 (1984).
73. R. L. Fork, C. H. Brito Cruz, P. C. Becker, and C. V. Shank, "Compression of optical pulses to six femtoseconds by using cubic phase compensation," *Opt. Lett.* **12**, 483-485 (1987).
74. U. Sadao, "Dispersion Compensation for a Femtosecond Cr:LiSAF Laser," *Japanese Journal of Applied Physics* **37**, 133 (1998).
75. P. Loiko and A. Major, "Dispersive properties of alexandrite and beryllium hexaaluminate crystals," *Opt. Mater. Express* **6**, 2177-2183 (2016).
76. F. Kaertner, "<https://ocw.mit.edu/courses/electrical-engineering-and-computer-science/6-977-ultrafast-optics-spring-2005/lecture-notes/>", retrieved 2018.
77. P. F. Moulton, "An Investigation of the Co:MgF₂ Laser System," *IEEE Journal of Quantum Electronics* **21**, 1582-1595 (1985).
78. Z. Chang, A. Rundquist, H. Wang, M. M. Murnane, and H. C. Kapteyn, "Generation of Coherent Soft X Rays at 2.7 nm Using High Harmonics," *Physical Review Letters* **79**, 2967-2970 (1997).
79. M. Drescher, M. Hentschel, R. Kienberger, M. Uiberacker, V. Yakovlev, A. Scrinzi, T. Westerwalbesloh, U. Kleineberg, U. Heinzmann, and F. Krausz, "Time-resolved atomic inner-shell spectroscopy," *Nature* **419**, 803 (2002).
80. G. J. Tearney, M. E. Brezinski, B. E. Bouma, S. A. Boppart, C. Pitris, J. F. Southern, and J. G. Fujimoto, "In Vivo Endoscopic Optical Biopsy with Optical Coherence Tomography," *Science* **276**, 2037 (1997).
81. D. Herriott, H. Kogelnik, and R. Kompfner, "Off-Axis Paths in Spherical Mirror Interferometers," *Appl. Opt.* **3**, 523-526 (1964).
82. A. Gerrard and J. M. Burch, *Introduction to matrix theory methods in optics* (DOVER PUBN Incorporated, 1975).
83. A. Sennaroglu, *Solid-State Lasers and Applications* (CRC Press, 2006).
84. A. Sennaroglu and J. G. Fujimoto, "Design criteria for Herriott-type multi-pass cavities for ultrashort pulse lasers," *Opt. Express* **11**, 1106-1113 (2003).
85. A. Sennaroglu, A. M. Kowalevich, E. P. Ippen, and J. G. Fujimoto, "Compact femtosecond lasers based on novel multi-pass cavities," *IEEE Journal of Quantum Electronics* **40**, 519-528 (2004).
86. X. Liu, D. Du, and G. Mourou, "Laser ablation and micromachining with ultrashort laser pulses," *IEEE Journal of Quantum Electronics* **33**, 1706-1716 (1997).
87. D. W. McCamant, P. Kukura, S. Yoon, and R. A. Mathies, "Femtosecond broadband stimulated Raman spectroscopy: Apparatus and methods," *Review of Scientific Instruments* **75**, 4971-4980 (2004).
88. H. Kneipp, J. Kolenda, P. Rairoux, B. Stein, D. Weidauer, J.-P. Wolf, and L. H. Woeste, "Ti:sapphire-laser-based lidar systems," in *Environmental Sensing '92*, (SPIE, 1992), 9.
89. M. D. Young, S. Backus, C. Durfee, and J. Squier, "Multiphoton imaging with a direct-diode pumped femtosecond Ti: sapphire laser," *Journal of microscopy* **249**, 83-86 (2013).
90. P. W. Roth, A. J. Maclean, D. Burns, and A. J. Kemp, "Directly diode-laser-pumped Ti:sapphire laser," *Opt. Lett.* **34**, 3334-3336 (2009).
91. R. Paschotta, "Noise of mode-locked lasers (Part II): timing jitter and other fluctuations," *Applied Physics B* **79**, 163-173 (2004).
92. J. Harrison, A. Finch, D. M. Rines, G. A. Rines, and P. F. Moulton, "Low-threshold, cw, all-solid-state Ti:Al₂O₃ laser," *Opt. Lett.* **16**, 581-583 (1991).

Bibliography

93. C. Grivas, C. Corbari, G. Brambilla, and P. G. Lagoudakis, "Tunable, continuous-wave Ti:sapphire channel waveguide lasers written by femtosecond and picosecond laser pulses," *Opt. Lett.* **37**, 4630-4632 (2012).
94. K. Gürel, V. J. Wittwer, M. Hoffmann, C. J. Saraceno, S. Hakobyan, B. Resan, A. Rohrbacher, K. Weingarten, S. Schilt, and T. Südmeyer, "Green-diode-pumped femtosecond Ti:Sapphire laser with up to 450 mW average power," *Opt. Express* **23**, 30043-30048 (2015).
95. S. Shota, H. Aruto, K. Hikaru, H. Kenichi, and K. Fumihiko, "Demonstration of a Ti:sapphire mode-locked laser pumped directly with a green diode laser," *Applied Physics Express* **7**, 022702 (2014).
96. P. W. Roth, D. Burns, and A. J. Kemp, "Power scaling of a directly diode-laser-pumped Ti:sapphire laser," *Opt. Express* **20**, 20629-20634 (2012).
97. U. Demirbas, S. Eggert, and A. Leitenstorfer, "Compact and efficient Cr:LiSAF lasers pumped by one single-spatial-mode diode: a minimal cost approach," *J. Opt. Soc. Am. B* **29**, 1894-1903 (2012).
98. P. Wagenblast, R. Ell, U. Morgner, F. Grawert, and F. X. Kärtner, "Diode-pumped 10-fs Cr³⁺:LiCAF laser," *Opt. Lett.* **28**, 1713-1715 (2003).
99. U. Sadao and T. Kenji, "Generation of 10 fs Pulses from a Diode-Pumped Kerr-Lens Mode-Locked Cr:LiSAF Laser," *Japanese Journal of Applied Physics* **39**, 3472 (2000).
100. V. Kubecek, R. Quintero-Torres, and J.-C. M. Diels, "Ultralow-pump-threshold laser diode pumped Cr:LiSAF laser," in *International Conference on Lasers, Applications, and Technologies 2002*, (SPIE, 2003), 5.
101. R. Scheps, J. F. Myers, H. B. Serreze, A. Rosenberg, R. C. Morris, and M. Long, "Diode-pumped Cr:LiSrAlF₆ laser," *Opt. Lett.* **16**, 820-822 (1991).
102. S. Tsuda, W. H. Knox, and S. T. Cundiff, "High efficiency diode pumping of a saturable Bragg reflector-mode-locked Cr:LiSAF femtosecond laser," *Applied Physics Letters* **69**, 1538-1540 (1996).
103. G. J. Valentine, J. M. Hopkins, P. Loza-Alvarez, G. T. Kennedy, W. Sibbett, D. Burns, and A. Valster, "Ultralow-pump-threshold, femtosecond Cr³⁺:LiSrAlF₆ laser pumped by a single narrow-stripe AlGaInP laser diode," *Opt. Lett.* **22**, 1639-1641 (1997).
104. B. Agate, B. Stormont, A. J. Kemp, C. T. A. Brown, U. Keller, and W. Sibbett, "Simplified cavity designs for efficient and compact femtosecond Cr:LiSAF lasers," *Optics Communications* **205**, 207-213 (2002).
105. D. Li, U. Demirbas, A. Benedick, A. Sennaroglu, J. G. Fujimoto, and F. X. Kärtner, "Attosecond timing jitter pulse trains from semiconductor saturable absorber mode-locked Cr:LiSAF lasers," *Opt. Express* **20**, 23422-23435 (2012).
106. D. Klimm, G. Lacayo, and P. Reiche, "Growth of Cr:LiCaAlF₆ and Cr:LiSrAlF₆ by the Czochralski method," *Journal of Crystal Growth* **210**, 683-693 (2000).
107. J. F. Pinto, L. Esterowitz, G. H. Rosenblatt, M. Kokta, and D. Peressini, "Improved Ti:sapphire laser performance with new high figure of merit crystals," *IEEE Journal of Quantum Electronics* **30**, 2612-2616 (1994).
108. L. K. Smith, S. A. Payne, W. L. Kway, L. L. Chase, and B. H. T. Chai, "Investigation of the Laser Properties of Cr³⁺:LiSrGaF₆," *Ieee Journal of Quantum Electronics* **28**, 2612-2618 (1992).
109. E. Sorokin, "Solid-State Materials for Few-Cycle Pulse Generation and Amplification," in *Few-Cycle Laser Pulse Generation and Its Applications*, F. X. Kärtner, ed. (Springer Berlin Heidelberg, Berlin, Heidelberg, 2004), pp. 3-73.
110. L. G. Deshazer, K. W. E. D. A. D. J. R. B. R. Kangas, and P. Liao, "Extended infrared operation of a titanium sapphire laser," in *Conference on Lasers and Electro-Optics*, OSA Technical Digest (Optical Society of America, 1987), THQ4.
111. W. Koechner, *Solid-state laser engineering*, 6th rev. and updated ed., Springer series in optical sciences, v. 1. (Springer, New York, 2006), pp. xvi, 747 p.
112. L J Atherton, a. S A Payne, and C. D. Brandle, "Oxide and Fluoride Laser Crystals," *Annual Review of Materials Science* **23**, 453-502 (1993).

Bibliography

113. C. Cihan, E. Beyatli, F. Canbaz, L. J. Chen, B. Sumpf, G. Erbert, A. Leitenstorfer, F. X. Kärtner, A. Sennaroglu, and U. Demirbas, "Gain-Matched Output Couplers for Efficient Kerr-Lens Mode-Locking of Low-Cost and High-Peak Power Cr:LiSAF Lasers," *IEEE Journal of Selected Topics in Quantum Electronics* **21**, 94-105 (2015).
114. A. Sanchez, R. E. Fahey, A. J. Strauss, and R. L. Aggarwal, "Room-temperature continuous-wave operation of a Ti:Al₂O₃ laser," *Opt. Lett.* **11**, 363-364 (1986).
115. W. R. E. D. C. L. Rapoport and A. Pinto, "Excited-State Absorption and Upconversion in Cr:LiSAF," in *Advanced Solid State Lasers*, OSA Proceedings Series (Optical Society of America, 1992), CL6.
116. M. A. Noginov, V. G. Ostroumov, I. A. Shcherbakov, V. A. Smirnov, D. A. E. D. G. Zubenko, and L. Chase, "Interaction of Excited Cr³⁺ Ions in Laser Crystals," in *Advanced Solid State Lasers*, OSA Proceedings Series (Optical Society of America, 1991), C3L6.
117. S. A. Payne, L. K. Smith, R. J. Beach, B. H. T. Chai, J. H. Tassano, L. D. DeLoach, W. L. Kway, R. W. Solarz, and W. F. Krupke, "Properties of Cr:LiSrAlF₆ crystals for laser operation," *Appl. Opt.* **33**, 5526-5536 (1994).
118. M. Stalder, M. Bass, and B. H. T. Chai, "Thermal quenching of fluorescence in chromium-doped fluoride laser crystals," *J. Opt. Soc. Am. B* **9**, 2271-2273 (1992).
119. H. S. Wang, P. Li Kam Wa, J. L. Lefaucheur, B. H. T. Chai, and A. Miller, "CW and self-mode-locking performance of a red pumped Cr³⁺: LiSr_{0.8}Ca_{0.2}AlF₆ laser," *Optics Communications* **110**, 679-688 (1994).
120. U. Sadao and M. Kenzo, "Thermal Characteristics of a Continuous-Wave Cr:LiSAF Laser," *Japanese Journal of Applied Physics* **36**, 4312 (1997).
121. F. Balembois, F. Druon, F. Falcoz, P. Georges, and A. Brun, "Performances of Cr:LiSrAlF₆ and Cr:LiSrGaF₆ for continuous-wave diode-pumped Q-switched operation," *Opt. Lett.* **22**, 387-389 (1997).
122. V. Pilla, T. Catunda, H. P. Jenssen, and A. Cassanho, "Fluorescence quantum efficiency measurements in the presence of Auger upconversion by the thermal lens method," *Opt. Lett.* **28**, 239-241 (2003).
123. V. Pilla, T. Catunda, S. Marcio Lima, A. Neto Medina, M. Luciano Baesso, H. P. Jenssen, and A. Cassanho, "Thermal quenching of the fluorescence quantum efficiency in colquiriite crystals measured by thermal lens spectrometry," *J. Opt. Soc. Am. B* **21**, 1784-1791 (2004).
124. S. A. Payne, L. L. Chase, and G. D. Wilke, "Optical spectroscopy of the new laser materials, LiSrAlF₆:Cr³⁺ and LiCaAlF₆:Cr³⁺," *Journal of Luminescence* **44**, 167-176 (1989).
125. F. Balembois, F. Falcoz, F. Kerboull, F. Druon, P. Georges, and A. Brun, "Theoretical and experimental investigations of small-signal gain for a diode-pumped Q-switched Cr:LiSAF laser," *IEEE Journal of Quantum Electronics* **33**, 269-278 (1997).
126. S. A. Payne, L. L. Chase, L. J. Atherton, J. A. Caird, W. L. Kway, M. D. Shinn, R. S. Hughes, and L. K. Smith, "Properties and performance of the LiCaAlF₆:Cr³⁺ laser material," in *OE/LASE '90*, (SPIE, 1990), 10.
127. P. Beaud, M. C. Richardson, Y. Chen, and B. H. T. Chai, "Optical amplification characteristics of Cr:LiSAF and Cr:LiCAF under flashlamp-pumping," *IEEE Journal of Quantum Electronics* **30**, 1259-1266 (1994).
128. B. W. Woods, S. A. Payne, J. E. Marion, R. S. Hughes, and L. E. Davis, "Thermomechanical and thermo-optical properties of the LiCaAlF₆:Cr³⁺ laser material," *J. Opt. Soc. Am. B* **8**, 970-977 (1991).
129. A. Sennaroglu, "Analysis and optimization of lifetime thermal loading in continuous-wave Cr⁴⁺-doped solid-state lasers," *J. Opt. Soc. Am. B* **18**, 1578-1586 (2001).
130. J. M. Eichenholz and M. Richardson, "Measurement of thermal lensing in Cr/sup 3+/-doped colquiriites," *IEEE Journal of Quantum Electronics* **34**, 910-919 (1998).
131. U. Demirbas, "Low-cost, highly efficient, and tunable ultrafast laser technology based on directly diode-pumped Cr:Colquiriites," (MIT, 2010).

132. J. K. Jabczyński, W. Zendzian, Z. Mierczyk, and Z. Frukacz, "Chromium-doped LiCAF laser passively Q switched with a V3+:YAG crystal," *Appl. Opt.* **40**, 6638-6645 (2001).
133. S. Tsuda, W. H. Knox, S. T. Cundiff, W. Y. Jan, and J. E. Cunningham, "Mode-locking ultrafast solid-state lasers with saturable Bragg reflectors," *IEEE Journal of Selected Topics in Quantum Electronics* **2**, 454-464 (1996).
134. U. Keller, K. J. Weingarten, F. X. Kartner, D. Kopf, B. Braun, I. D. Jung, R. Fluck, C. Honninger, N. Matuschek, and J. A. d. Au, "Semiconductor saturable absorber mirrors (SESAM's) for femtosecond to nanosecond pulse generation in solid-state lasers," *IEEE Journal of Selected Topics in Quantum Electronics* **2**, 435-453 (1996).
135. A. J. Kemp, B. Stormont, B. Agate, C. T. A. Brown, U. Keller, and W. Sibbett, "Gigahertz repetition-rate from directly diode-pumped femtosecond Cr:LiSAF laser," in *Electronics Letters*, (Institution of Engineering and Technology, 2001), pp. 1457-1458.
136. J. M. Hopkins, G. J. Valentine, B. Agate, A. J. Kemp, U. Keller, and W. Sibbett, "Highly compact and efficient femtosecond Cr:LiSAF lasers," *IEEE Journal of Quantum Electronics* **38**, 360-368 (2002).
137. U. Demirbas, G. S. Petrich, D. Li, A. Sennaroglu, L. A. Kolodziejski, F. X. Kärtner, and J. G. Fujimoto, "Femtosecond tuning of Cr:colquiriite lasers with AlGaAs-based saturable Bragg reflectors," *J. Opt. Soc. Am. B* **28**, 986-993 (2011).
138. D. Kopf, A. Prasad, G. Zhang, M. Moser, and U. Keller, "Broadly tunable femtosecond Cr:LiSAF laser," *Opt. Lett.* **22**, 621-623 (1997).
139. I. T. Sorokina, E. Sorokin, E. Wintner, A. Cassanho, H. P. Jenssen, and R. Szipocs, "14-fs pulse generation in Kerr-lens mode-locked prismless Cr:LiSGaF and Cr:LiSAF lasers: observation of pulse self-frequency shift," *Opt. Lett.* **22**, 1716-1718 (1997).
140. R. Mellish, S. C. W. Hyde, N. P. Barry, R. Jones, P. M. W. French, J. R. Taylor, C. J. van der Poel, and A. Valster, "All-solid-state diode-pumped Cr:LiSAF femtosecond oscillator and regenerative amplifier," *Applied Physics B* **65**, 221-226 (1997).
141. A. Robertson, R. Knappe, and R. Wallenstein, "Diode-pumped broadly tunable (809–910 nm) femtosecond Cr:LiSAF laser," *Optics Communications* **147**, 294-298 (1998).
142. E. Sorokin, "Solid-state materials for few-cycle pulse generation and amplification," in *Few-cycle laser pulse generation and its applications*, F. X. Kartner, ed. (Springer-Verlag, Berlin, 2004), pp. 3-71.
143. S. Uemura and K. Torizuka, "Generation of 12-fs pulses from a diode-pumped Kerr-lens mode-locked Cr:LiSAF laser," *Opt. Lett.* **24**, 780-782 (1999).
144. S. Uemura and K. Miyazaki, "Femtosecond Cr:LiSAF laser pumped by a single diode laser," *Optics Communications* **138**, 330-332 (1997).
145. L.-J. Chen, M. Y. Sander, and F. X. Kärtner, "Kerr-lens mode locking with minimum nonlinearity using gain-matched output couplers," *Opt. Lett.* **35**, 2916-2918 (2010).
146. F. Canbaz, E. Beyatli, L.-J. Chen, A. Sennaroglu, F. X. Kärtner, and U. Demirbas, "Highly efficient and robust operation of Kerr-lens mode-locked Cr:LiSAF lasers using gain-matched output couplers," *Opt. Lett.* **39**, 327-330 (2014).
147. B. Sumpf, P. Adamiec, M. Zorn, H. Wenzel, and G. Erbert, "Nearly Diffraction-Limited Tapered Lasers at 675 nm With 1-W Output Power and Conversion Efficiencies Above 30%," *Ieee Photonics Technology Letters* **23**, 266-268 (2011).
148. U. Demirbas, M. Schmalz, B. Sumpf, G. Erbert, G. S. Petrich, L. A. Kolodziejski, J. G. Fujimoto, F. X. Kärtner, and A. Leitenstorfer, "Femtosecond Cr:LiSAF and Cr:LiCAF lasers pumped by tapered diode lasers," *Opt. Express* **19**, 20444-20461 (2011).
149. N. P. Barry, S. C. W. Hyde, R. Mellish, P. M. W. French, J. R. Taylor, C. J. v. d. Poel, and A. Valster, "All-solid-state femtosecond diode-pumped Cr:LiSAF regenerative amplifier," *Electronics Letters* **30**, 1761-1762 (1994).
150. A. Agnesi, F. Pirzio, E. Ugolotti, S. Y. Choi, D. Yeom, II, and F. Rotermund, "Femtosecond single-mode diode-pumped Cr:LiSAF laser mode-locked with single-walled carbon nanotubes," *Optics Communications* **285**, 742-745 (2012).

151. U. Demirbas, G. S. Petrich, S. Nabanja, J. R. Birge, L. A. Kolodziejski, F. X. Kärtner, and J. G. Fujimoto, "Widely-tunable femtosecond operation of Cr:LiSAF lasers using broadband saturable Bragg reflectors," in *CLEO/QELS: 2010 Laser Science to Photonic Applications*, (2010), 1-2.
152. G. V. Bukin, S. Y. Volkov, V. N. Matrosov, B. K. Sevast'yanov, and M. I. Timoshechkin, "Stimulated emission from alexandrite ($\text{BeAl}_2\text{O}_4:\text{Cr}^{3+}$)," *Soviet Journal of Quantum Electronics* **8**, 671 (1978).
153. F. E. F. and F. J. H., "Flux Growth of Chrysoberyl and Alexandrite," *Journal of the American Ceramic Society* **47**, 274-276 (1964).
154. R. C. Morris and C. F. Cline, "Chromium-doped beryllium aluminate lasers," (Google Patents, 1976).
155. J. C. Walling, "Tunable Cw Alexandrite Lasers," *Journal of the Optical Society of America* **69**, 1436-1436 (1979).
156. J. Walling, O. Peterson, H. Jenssen, R. Morris, and E. O. Dell, "Tunable alexandrite lasers," *IEEE Journal of Quantum Electronics* **16**, 1302-1315 (1980).
157. J. C. Walling, D. F. Heller, H. Samelson, D. J. Harter, J. A. Pete, and R. C. Morris, "Tunable alexandrite lasers: development and performance," *IEEE Journal of Quantum Electronics* **QE-21**, 1568-1581 (1985).
158. W. Koechner, *Solid state laser engineering* (Springer-Verlag, 1976).
159. W. Gadomski, B. Ratajska-Gadomska, and R. Meucci, "Homoclinic dynamics of the vibronic laser," *Chaos, Solitons & Fractals* **17**, 387-396 (2003).
160. E. Beyatli, I. Baali, B. Sumpf, G. Erbert, A. Leitenstorfer, A. Sennaroglu, and U. Demirbas, "Tapered diode-pumped continuous-wave alexandrite laser," *J. Opt. Soc. Am. B* **30**, 3184-3192 (2013).
161. I. Yorulmaz, E. Beyatli, A. Kurt, A. Sennaroglu, and U. Demirbas, "Efficient and low-threshold Alexandrite laser pumped by a single-mode diode," *Opt. Mater. Express* **4**, 776-789 (2014).
162. P. Pichon, A. Barbet, J. P. Blanchot, F. Druon, F. Balembois, and P. Georges, "LED-pumped alexandrite laser oscillator and amplifier," *Opt Lett* **42**, 4191-4194 (2017).
163. S. Guch and C. E. Jones, "Alexandrite-laser performance at high temperature," *Opt. Lett.* **7**, 608-610 (1982).
164. H. Samelson, J. C. Walling, and D. F. Heller, "Unique Applications Of Alexandrite Lasers," in *1982 Technical Symposium East*, (SPIE, 1983), 10.
165. S. Liu, J. Liu, and L. Wang, "High output energy tunable alexandrite laser," *Chin. Opt. Lett.* **5**, S16-S18 (2007).
166. P. Bakule, P. E. G. Baird, M. G. Boshier, S. L. Cornish, D. F. Heller, K. Jungmann, I. C. Lane, V. Meyer, P. H. G. Sandars, W. T. Toner, M. Towrie, and J. C. Walling, "A chirp-compensated, injection-seeded alexandrite laser," *Applied Physics B* **71**, 11-17 (2000).
167. K. Torizuka, M. Yamashita, and T. Yabiku, "Continuous-wave alexandrite laser pumped by a direct-current mercury arc lamp," *Appl. Opt.* **32**, 7394-7398 (1993).
168. A. Munk, B. Jungbluth, M. Strotkamp, H. D. Hoffmann, R. Poprawe, J. Höffner, and F. J. Lübken, "Diode-pumped alexandrite ring laser in single-longitudinal mode operation for atmospheric lidar measurements," *Opt. Express* **26**, 14928-14935 (2018).
169. G. M. Thomas, A. Minassian, X. Sheng, and M. J. Damzen, "Diode-pumped Alexandrite lasers in Q-switched and cavity-dumped Q-switched operation," *Opt. Express* **24**, 27212-27224 (2016).
170. R. Scheps, J. F. Myers, T. R. Glesne, and H. B. Serreze, "Monochromatic end-pumped operation of an alexandrite laser," *Optics Communications* **97**, 363-366 (1993).
171. A. Munk, B. Jungbluth, M. Strotkamp, H.-D. Hoffmann, R. Poprawe, and J. Höffner, "Diode-pumped Alexandrite ring laser for lidar applications," in *SPIE LASE*, (SPIE, 2016), 6.
172. P. Pichon, A. Barbet, J.-P. Blanchot, F. Druon, F. Balembois, and P. Georges, "LED-pumped alexandrite laser oscillator and amplifier," *Opt. Lett.* **42**, 4191-4194 (2017).

Bibliography

173. J. W. Kuper and D. C. Brown, "High efficiency CW green-pumped alexandrite lasers," in 2006), 61000T-61000T-61008.
174. G. Shirin and M. Arkady, "High power continuous-wave Alexandrite laser with green pump," *Laser Physics* **26**, 075001 (2016).
175. J. W. Kuper, T. Chin, and H. E. E. D. D. G. Aschoff, "Extended Tuning Range of Alexandrite at Elevated Temperatures," in *Advanced Solid State Lasers*, OSA Proceedings Series (Optical Society of America, 1990), CL3.
176. L. G. DeShazer and K. W. Kangas, "Extended infrared operation of titanium sapphire laser," *Conference on Lasers and Electro Optics* **14**, 296-298 (1987).
177. J. Walling, D. Heller, H. Samelson, D. Harter, J. Pete, and R. Morris, "Tunable alexandrite lasers: Development and performance," *IEEE Journal of Quantum Electronics* **21**, 1568-1581 (1985).
178. A. Teppitaksak, A. Minassian, G. M. Thomas, and M. J. Damzen, "High efficiency >26 W diode end-pumped Alexandrite laser," *Opt. Express* **22**, 16386-16392 (2014).
179. M. J. Damzen, G. M. Thomas, and A. Minassian, "Multi-watt diode-pumped alexandrite laser operation," in *CLEO Europe*, (Optical Society of America, Munich, 2013).
180. A. Hariharan, M. E. Fermann, M. L. Stock, D. J. Harter, and J. Squier, "Alexandrite-pumped alexandrite regenerative amplifier for femtosecond pulse amplification," *Opt. Lett.* **21**, 128-130 (1996).
181. D. J. Harter and P. Bado, "Wavelength tunable alexandrite regenerative amplifier," *Appl. Opt.* **27**, 4392-4395 (1988).
182. M. Pessot, J. Squier, P. Bado, G. Mourou, and D. J. Harter, "Chirped pulse amplification of 300 fs pulses in an alexandrite regenerative amplifier," *IEEE Journal of Quantum Electronics* **25**, 61-66 (1989).
183. J. C. Walling, D. F. Heller, H. Samelson, D. J. Harter, J. A. Pete, and R. C. Morris, "Tunable Alexandrite Lasers - Development and Performance," *IEEE Journal of Quantum Electronics* **21**, 1568-1581 (1985).
184. Y. J. Lee, H. J. Shin, T.-K. Noh, K.-H. Choi, and S.-E. Chang, "Treatment of Melasma and Post-Inflammatory Hyperpigmentation by a Picosecond 755-nm Alexandrite Laser in Asian Patients," *Annals of Dermatology* **29**, 779-781 (2017).
185. C. Dierickx, "Using normal and high pulse coverage with picosecond laser treatment of wrinkles and acne scarring: Long term clinical observations," *Lasers Surg Med* (2017).
186. R. R. Anderson, "Lasers for dermatology and skin biology," *J Invest Dermatol* **133**, E21-23 (2013).
187. M. J. Damzen, G. M. Thomas, A. Teppitaksak, and A. Minassian, "Progress in diode-pumped alexandrite lasers as a new resource for future space lidar missions," in *International Conference on Space Optics — ICSO 2014*, (SPIE, 2017), 9.
188. A. T. Coney, G. M. Thomas, A. Minassian, and M. J. Damzen, "High-energy diode-pumped alexandrite laser development for remote sensing," in *2017 Conference on Lasers and Electro-Optics Europe & European Quantum Electronics Conference (CLEO/Europe-EQEC)*, (2017), 1-1.
189. M. Strotkamp, U. Witte, A. Munk, A. Hartung, S. Gausmann, S. Hengesbach, M. Traub, H.-D. Hoffmann, J. Hoeffner, and B. Jungbluth, "Broadly tunable, longitudinally diode-pumped Alexandrite laser," in *SPIE LASE*, (SPIE, 2014), 6.
190. V. Wulfmeyer, J. Bösenberg, S. Lehmann, C. Senff, and S. Schmitz, "Injection-seeded alexandrite ring laser: performance and application in a water-vapor differential absorption lidar," *Opt. Lett.* **20**, 638-640 (1995).
191. J. A. McKay and T. D. Wilkerson, "Laser source for space-flight elastic backscatter, differential absorption, and wind speed Doppler lidar," in *SPIE's 1996 International Symposium on Optical Science, Engineering, and Instrumentation*, (SPIE, 1996), 13.
192. R. M. Williams, W. R. Zipfel, and W. W. Webb, "Multiphoton microscopy in biological research," *Curr Opin Chem Biol* **5**, 603-608 (2001).
193. W. R. Zipfel, R. M. Williams, and W. W. Webb, "Nonlinear magic: multiphoton microscopy in the biosciences," *Nat Biotechnol* **21**, 1369-1377 (2003).

194. S. Huang, A. A. Heikal, and W. W. Webb, "Two-photon fluorescence spectroscopy and microscopy of NAD(P)H and flavoprotein," *Biophys J* **82**, 2811-2825 (2002).
195. M. Schneider, S. Barozzi, I. Testa, M. Faretta, and A. Diaspro, "Two-Photon Activation and Excitation Properties of PA-GFP in the 720–920-nm Region," *Biophysical Journal* **89**, 1346-1352 (2005).
196. S. Sakadzic, U. Demirbas, T. R. Mempel, A. Moore, S. Ruvinskaya, D. A. Boas, A. Sennaroglu, F. X. Kartner, and J. G. Fujimoto, "Multi-photon microscopy with a low-cost and highly efficient Cr:LiCAF laser," *Opt. Express* **16**, 20848-20863 (2008).
197. S. Imai and H. Ito, "Long-pulse ultraviolet-laser sources based on tunable alexandrite lasers," *IEEE Journal of Quantum Electronics* **34**, 573-576 (1998).
198. "Third-harmonic generation of an alexandrite laser in β -BaB₂O₄," *Applied Physics Letters* **54**, 1206-1208 (1989).
199. J. W. Kuper and D. C. Brown, "Highly efficient CW green pumped Alexandrite Lasers," in *Solid State Lasers XV: Technology and Devices*, (SPIE, 2006),
200. J. C. Walling, O. G. Peterson, and R. C. Morris, "Tunable Cw Alexandrite Laser," *IEEE Journal of Quantum Electronics* **16**, 120-121 (1980).
201. M. Fibrich, J. Šulc, D. Vyhřídál, H. Jelínková, and M. Čech, "Alexandrite spectroscopic and laser characteristic investigation within a 78–400 K temperature range," *Laser Physics* **27**, 115801 (2017).
202. V. N. Lisitsyn, V. N. Matsorov, V. P. Orekhova, E. P. Pestryakov, B. K. Sevast'yanov, V. I. Trunov, V. N. Zenin, and Y. L. Remigailo, "Generation of 0.7-0.8 picosecond pulses in an alexandrite laser with passive mode locking," *Soviet Journal of Quantum Electronics* **vol.12**, 368-370 (1982).
203. D. Bruneau, T. A. d. Lions, P. Quaglia, and J. Pelon, "Injection-seeded pulsed alexandrite laser for differential absorption lidar application," *Appl. Opt.* **33**, 3941-3950 (1994).
204. U. Parali, X. Sheng, A. Minassian, G. Tawy, J. Sathian, G. M. Thomas, and M. J. Damzen, "Diode-pumped Alexandrite laser with passive SESAM Q-switching and wavelength tunability," *Optics Communications* **410**, 970-976 (2018).
205. F. Völker, Q. Lü, and H. Weber, "Passive mode-locking of an Alexandrite laser for picosecond pulse generation," *Journal of Applied Physics* **69**, 3432-3439 (1991).
206. S. Ghanbari, R. Akbari, and A. Major, "Femtosecond Kerr-lens mode-locked Alexandrite laser," *Opt. Express* **24**, 14836-14840 (2016).
207. S. Ghanbari, K. A. Fedorova, A. B. Krysa, E. U. Rafailov, and A. Major, "Femtosecond Alexandrite laser passively mode-locked by an InP/InGaP quantum-dot saturable absorber," *Opt. Lett.* **43**, 232-234 (2018).
208. W. Gadomski and B. Ratajska-Gadomska, "Homoclinic orbits and chaos in the vibronic short-cavity standing-wave alexandrite laser," *J. Opt. Soc. Am. B* **17**, 188-197 (2000).
209. B. H. Soffer, "Giant Pulse Laser Operation by a Passive, Reversibly Bleachable Absorber," *Journal of Applied Physics* **35**, 2551-2551 (1964).
210. G. Bret and F. Gires, "GIANT-PULSE LASER AND LIGHT AMPLIFIER USING VARIABLE TRANSMISSION COEFFICIENT GLASSES AS LIGHT SWITCHES," *Applied Physics Letters* **4**, 175-176 (1964).
211. P. T. Guerreiro, "PbS quantum-dot doped glasses as saturable absorbers for mode locking of a Cr:forsterite laser," *Applied Physics Letters* **71**, 1595-1597 (1997).
212. S. Y. Set, H. Yaguchi, Y. Tanaka, and M. Jablonski, "Ultrafast fiber pulsed lasers incorporating carbon nanotubes," *IEEE Journal of Selected Topics in Quantum Electronics* **10**, 137-146 (2004).
213. T. Hasan, Z. Sun, F. Wang, F. Bonaccorso, P. H. Tan, A. G. Rozhin, and A. C. Ferrari, "Nanotube–Polymer Composites for Ultrafast Photonics," *Advanced Materials* **21**, 3874-3899 (2009).
214. R. Woodward and E. Kelleher, "2D Saturable Absorbers for Fibre Lasers," *Applied Sciences* **5**, 1440 (2015).
215. G. Ruess and F. Vogt, "Höchstlamellarer Kohlenstoff aus Graphitoxhydroxyd," *Monatshefte für Chemie und verwandte Teile anderer Wissenschaften* **78**, 222-242 (1948).

216. C. Schafhaeuti, "LXXXVI. On the combinations of carbon with silicon and iron, and other metals, forming the different species of cast iron, steel, and malleable iron," *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science* **16**, 570-590 (1840).
217. H.-P. Boehm and E. Stumpp, "Citation errors concerning the first report on exfoliated graphite," *Carbon* **45**, 1381-1383 (2007).
218. H. P. Boehm, "Graphene—How a Laboratory Curiosity Suddenly Became Extremely Interesting," *Angewandte Chemie International Edition* **49**, 9332-9335 (2010).
219. F. Lincoln Vogel, "The electrical conductivity of graphite intercalated with superacid fluorides: experiments with antimony pentafluoride," *Journal of Materials Science* **12**, 982-986 (1977).
220. A. Hamwi, S. Mouras, D. Djurado, and J. C. Cousseins, "New synthesis of first stage graphite intercalation compounds with fluorides," *Journal of Fluorine Chemistry* **35**, 151 (1987).
221. H. P. Boehm, R. Setton, and E. Stumpp, "Nomenclature and terminology of graphite intercalation compounds (IUPAC Recommendations 1994)," in *Pure and Applied Chemistry*, (1994), p. 1893.
222. P. R. Wallace, "The Band Theory of Graphite," *Physical Review* **71**, 622-634 (1947).
223. K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, and A. A. Firsov, "Electric Field Effect in Atomically Thin Carbon Films," *Science* **306**, 666-669 (2004).
224. A. K. Geim and K. S. Novoselov, "The rise of graphene," *Nature Materials* **6**, 183 (2007).
225. A. H. Castro Neto, F. Guinea, N. M. R. Peres, K. S. Novoselov, and A. K. Geim, "The electronic properties of graphene," *Reviews of Modern Physics* **81**, 109-162 (2009).
226. K. I. Bolotin, K. J. Sikes, Z. Jiang, M. Klima, G. Fudenberg, J. Hone, P. Kim, and H. L. Stormer, "Ultrahigh electron mobility in suspended graphene," *Solid State Communications* **146**, 351-355 (2008).
227. A. S. Mayorov, R. V. Gorbachev, S. V. Morozov, L. Britnell, R. Jalil, L. A. Ponomarenko, P. Blake, K. S. Novoselov, K. Watanabe, T. Taniguchi, and A. K. Geim, "Micrometer-Scale Ballistic Transport in Encapsulated Graphene at Room Temperature," *Nano Letters* **11**, 2396-2399 (2011).
228. A. A. Balandin, S. Ghosh, W. Bao, I. Calizo, D. Teweldebrhan, F. Miao, and C. N. Lau, "Superior Thermal Conductivity of Single-Layer Graphene," *Nano Letters* **8**, 902-907 (2008).
229. J. Yu, G. Liu, A. V. Sumant, V. Goyal, and A. A. Balandin, "Graphene-on-Diamond Devices with Increased Current-Carrying Capacity: Carbon sp²-on-sp³ Technology," *Nano Letters* **12**, 1603-1608 (2012).
230. A. Roberts, D. Cormode, C. Reynolds, T. Newhouse-Illige, B. J. Leroy, and A. S. Sandhu, "Response of graphene to femtosecond high-intensity laser irradiation," *Applied Physics Letters* **99**(2011).
231. M. Liu, X. Yin, E. Ulin-Avila, B. Geng, T. Zentgraf, L. Ju, F. Wang, and X. Zhang, "A graphene-based broadband optical modulator," *Nature* **474**, 64 (2011).
232. M. Liu, X. Yin, and X. Zhang, "Double-Layer Graphene Optical Modulator," *Nano Letters* **12**, 1482-1485 (2012).
233. H. Yingtao, P. Marianna, C. Joris, B. Steven, A. Inge, H. Cedric, A. Philippe, and T. Dries, "Broadband 10 Gb/s operation of graphene electro-absorption modulator on silicon," *Laser & Photonics Reviews* **10**, 307-316 (2016).
234. C. T. Phare, Y.-H. Daniel Lee, J. Cardenas, and M. Lipson, "Graphene electro-optic modulator with 30 GHz bandwidth," *Nature Photonics* **9**, 511 (2015).
235. I. Goykhman, U. Sassi, B. Desiatov, N. Mazurski, S. Milana, D. de Fazio, A. Eiden, J. Khurgin, J. Shappir, U. Levy, and A. C. Ferrari, "On-Chip Integrated, Silicon-Graphene Plasmonic Schottky Photodetector with High Responsivity and Avalanche Photogain," *Nano Letters* **16**, 3005-3013 (2016).
236. D. Spirito, D. Coquillat, S. L. D. Bonis, A. Lombardo, M. Bruna, A. C. Ferrari, V. Pellegrini, A. Tredicucci, W. Knap, and M. S. Vitiello, "High performance bilayer-graphene terahertz detectors," *Applied Physics Letters* **104**, 061111 (2014).

237. L. Liao, Y.-C. Lin, M. Bao, R. Cheng, J. Bai, Y. Liu, Y. Qu, K. L. Wang, Y. Huang, and X. Duan, "High-speed graphene transistors with a self-aligned nanowire gate," *Nature* **467**, 305 (2010).
238. A. C. Ferrari, F. Bonaccorso, V. Fal'ko, K. S. Novoselov, S. Roche, P. Bøggild, S. Borini, F. H. L. Koppens, V. Palermo, N. Pugno, J. A. Garrido, R. Sordan, A. Bianco, L. Ballerini, M. Prato, E. Lidorikis, J. Kivioja, C. Marinelli, T. Ryhänen, A. Morpurgo, J. N. Coleman, V. Nicolosi, L. Colombo, A. Fert, M. Garcia-Hernandez, A. Bachtold, G. F. Schneider, F. Guinea, C. Dekker, M. Barbone, Z. Sun, C. Galiotis, A. N. Grigorenko, G. Konstantatos, A. Kis, M. Katsnelson, L. Vandersypen, A. Loiseau, V. Morandi, D. Neumaier, E. Treossi, V. Pellegrini, M. Polini, A. Tredicucci, G. M. Williams, B. Hee Hong, J.-H. Ahn, J. Min Kim, H. Zirath, B. J. van Wees, H. van der Zant, L. Occhipinti, A. Di Matteo, I. A. Kinloch, T. Seyller, E. Quesnel, X. Feng, K. Teo, N. Rupesinghe, P. Hakonen, S. R. T. Neil, Q. Tannock, T. Löfwander, and J. Kinaret, "Science and technology roadmap for graphene, related two-dimensional crystals, and hybrid systems," *Nanoscale* **7**, 4598-4810 (2015).
239. M. Dragoman, A. A. Muller, D. Dragoman, F. Coccetti, and R. Plana, "Terahertz antenna based on graphene," *Journal of Applied Physics* **107**, 104313 (2010).
240. H. Wang, A. Hsu, J. Wu, J. Kong, and T. Palacios, "Graphene-Based Ambipolar RF Mixers," *IEEE Electron Device Letters* **31**, 906-908 (2010).
241. S. A. Awan, A. Lombardo, A. Colli, G. Privitera, T. S. Kulmala, J. M. Kivioja, M. Koshino, and A. C. Ferrari, "Transport conductivity of graphene at RF and microwave frequencies," *2D Materials* **3**, 015010 (2016).
242. G. Liang, N. Neophytou, D. E. Nikonov, and M. S. Lundstrom, "Performance Projections for Ballistic Graphene Nanoribbon Field-Effect Transistors," *IEEE Transactions on Electron Devices* **54**, 677-682 (2007).
243. J. R. Williams, L. DiCarlo, and C. M. Marcus, "Quantum Hall Effect in a Gate-Controlled p-n Junction of Graphene," *Science* **317**, 638-641 (2007).
244. K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, M. I. Katsnelson, I. V. Grigorieva, S. V. Dubonos, and A. A. Firsov, "Two-dimensional gas of massless Dirac fermions in graphene," *Nature* **438**, 197 (2005).
245. D. Brida, A. Tomadin, C. Manzoni, Y. J. Kim, A. Lombardo, S. Milana, R. R. Nair, K. S. Novoselov, A. C. Ferrari, G. Cerullo, and M. Polini, "Ultrafast collinear scattering and carrier multiplication in graphene," *Nature Communications* **4**, 1987 (2013).
246. M. Breusing, C. Ropers, and T. Elsaesser, "Ultrafast Carrier Dynamics in Graphite," *Physical Review Letters* **102**, 086809 (2009).
247. D. Sun, Z.-K. Wu, C. Divin, X. Li, C. Berger, W. A. de Heer, P. N. First, and T. B. Norris, "Ultrafast Relaxation of Excited Dirac Fermions in Epitaxial Graphene Using Optical Differential Transmission Spectroscopy," *Physical Review Letters* **101**, 157402 (2008).
248. R. R. Nair, P. Blake, A. N. Grigorenko, K. S. Novoselov, T. J. Booth, T. Stauber, N. M. R. Peres, and A. K. Geim, "Fine Structure Constant Defines Visual Transparency of Graphene," *Science* **320**, 1308 (2008).
249. C. Casiraghi, A. Hartschuh, E. Lidorikis, H. Qian, H. Harutyunyan, T. Gokus, K. S. Novoselov, and A. C. Ferrari, "Rayleigh Imaging of Graphene and Graphene Layers," *Nano Letters* **7**, 2711-2717 (2007).
250. Z. Sun, T. Hasan, F. Torrisi, D. Popa, G. Privitera, F. Wang, F. Bonaccorso, D. M. Basko, and A. C. Ferrari, "Graphene Mode-Locked Ultrafast Laser," *ACS Nano* **4**, 803-810 (2010).
251. H. Tawfique, S. Zhipei, W. Fengqiu, B. Francesco, T. P. Heng, R. A. G., and F. A. C., "Nanotube-Polymer Composites for Ultrafast Photonics," *Advanced Materials* **21**, 3874-3899 (2009).
252. J. M. Dawlaty, S. Shivaraman, M. Chandrashekhhar, F. Rana, and M. G. Spencer, "Measurement of ultrafast carrier dynamics in epitaxial graphene," *Applied Physics Letters* **92**, 042116 (2008).
253. Q. Bao, H. Zhang, Y. Wang, Z. Ni, Y. Yan, Z. X. Shen, K. P. Loh, and D. Y. Tang, "Atomic-Layer Graphene as a Saturable Absorber for Ultrafast Pulsed Lasers," *Advanced Functional Materials* **19**, 3077-3083 (2009).

254. J. E. Proctor, D. M. Armada, and A. Vijayaraghavan, *An Introduction to Graphene and Carbon Nanotubes* (CRC Press, 2017).
255. J. C. Slonczewski and P. R. Weiss, "Band Structure of Graphite," *Physical Review* **109**, 272-279 (1958).
256. W. B. Cho, J. W. Kim, H. W. Lee, S. Bae, B. H. Hong, S. Y. Choi, I. H. Baek, K. Kim, D.-I. Yeom, and F. Rotermund, "High-quality, large-area monolayer graphene for efficient bulk laser mode-locking near 1.25 μm ," *Opt. Lett.* **36**, 4089-4091 (2011).
257. J. M. Dawlaty, S. Shivaraman, J. Strait, P. George, M. Chandrashekar, F. Rana, M. G. Spencer, D. Veksler, and Y. Chen, "Measurement of the optical absorption spectra of epitaxial graphene from terahertz to visible," *Applied Physics Letters* **93**, 131905 (2008).
258. N. M. R. Peres, F. Guinea, and A. H. Castro Neto, "Electronic properties of disordered two-dimensional carbon," *Physical Review B* **73**, 125411 (2006).
259. V. P. Gusynin, S. G. Sharapov, and J. P. Carbotte, "Magnetooptical conductivity in graphene," *Journal of Physics: Condensed Matter* **19**, 026222 (2007).
260. S. Das Sarma, S. Adam, E. H. Hwang, and E. Rossi, "Electronic transport in two-dimensional graphene," *Reviews of Modern Physics* **83**, 407-470 (2011).
261. N. M. R. Peres, "Colloquium: The transport properties of graphene: An introduction," *Reviews of Modern Physics* **82**, 2673-2700 (2010).
262. J. Horng, C.-F. Chen, B. Geng, C. Girit, Y. Zhang, Z. Hao, H. A. Bechtel, M. Martin, A. Zettl, M. F. Crommie, Y. R. Shen, and F. Wang, "Drude conductivity of Dirac fermions in graphene," *Physical Review B* **83**, 165113 (2011).
263. K. F. Mak, L. Ju, F. Wang, and T. F. Heinz, "Optical spectroscopy of graphene: From the far infrared to the ultraviolet," *Solid State Communications* **152**, 1341-1349 (2012).
264. M. M. Leandro, M. Kin Fai, A. H. C. Neto, N. M. R. Peres, and F. H. Tony, "Observation of intra- and inter-band transitions in the transient optical response of graphene," *New Journal of Physics* **15**, 015009 (2013).
265. T. Stauber, N. M. R. Peres, and A. K. Geim, "Optical conductivity of graphene in the visible region of the spectrum," *Physical Review B* **78**, 085432 (2008).
266. K. F. Mak, M. Y. Sfeir, Y. Wu, C. H. Lui, J. A. Misewich, and T. F. Heinz, "Measurement of the Optical Conductivity of Graphene," *Physical Review Letters* **101**, 196405 (2008).
267. Z. Q. Li, E. A. Henriksen, Z. Jiang, Z. Hao, M. C. Martin, P. Kim, H. L. Stormer, and D. N. Basov, "Dirac charge dynamics in graphene by infrared spectroscopy," *Nature Physics* **4**, 532 (2008).
268. F. Wang, Y. Zhang, C. Tian, C. Girit, A. Zettl, M. Crommie, and Y. R. Shen, "Gate-Variable Optical Transitions in Graphene," *Science* **320**, 206 (2008).
269. I. Baylam, S. Ozharar, N. Kakenov, C. Kocabas, and A. Sennaroglu, "Femtosecond pulse generation from a Ti³⁺:sapphire laser near 800nm with voltage reconfigurable graphene saturable absorbers," *Opt. Lett.* **42**, 1404-1407 (2017).
270. E. Garmire, "Resonant optical nonlinearities in semiconductors," *IEEE Journal of Selected Topics in Quantum Electronics* **6**, 1094-1110 (2000).
271. F. Zhang, S. Han, Y. Liu, Z. Wang, and X. Xu, "Dependence of the saturable absorption of graphene upon excitation photon energy," *Applied Physics Letters* **106**, 091102 (2015).
272. Q. Bao, "Atomic-Layer Graphene as a Saturable Absorber for Ultrafast Pulsed Lasers," *Advanced Functional Materials* **19**, 3077-3083 (2009).
273. B. A. Ruzicka, S. Wang, L. K. Werake, B. Weintrub, K. P. Loh, and H. Zhao, "Hot carrier diffusion in graphene," *Physical Review B* **82**, 195414 (2010).
274. I. H. Baek, H. W. Lee, S. Bae, B. H. Hong, Y. H. Ahn, D.-I. Yeom, and F. Rotermund, "Efficient Mode-Locking of Sub-70-fs Ti:Sapphire Laser by Graphene Saturable Absorber," *Applied Physics Express* **5**, 032701 (2012).
275. F. Canbaz, N. Kakenov, C. Kocabas, U. Demirbas, and A. Sennaroglu, "Generation of sub-20-fs pulses from a graphene mode-locked laser," *Opt. Express* **25**, 2834-2839 (2017).

276. S. Davide Di Dio Cafiso, E. Ugolotti, A. Schmidt, V. Petrov, U. Griebner, A. Agnesi, W. B. Cho, B. H. Jung, F. Rotermund, S. Bae, B. H. Hong, G. Reali, and F. Pirzio, "Sub-100-fs Cr:YAG laser mode-locked by monolayer graphene saturable absorber," *Opt. Lett.* **38**, 1745-1747 (2013).
277. M. N. Cizmeciyan, J. W. Kim, S. Bae, B. H. Hong, F. Rotermund, and A. Sennaroglu, "Graphene mode-locked femtosecond Cr:ZnSe laser at 2500nm," *Opt. Lett.* **38**, 341-343 (2013).
278. G. Zhu, X. Zhu, F. Wang, S. Xu, Y. Li, X. Guo, K. Balakrishnan, R. A. Norwood, and N. Peyghambarian, "Graphene Mode-Locked Fiber Laser at 2.8 μm ," *IEEE Photonics Technology Letters* **28**, 7-10 (2016).
279. G. Zhu, X. Zhu, K. Balakrishnan, R. A. Norwood, and N. Peyghambarian, "Fe²⁺:ZnSe and graphene Q-switched singly Ho³⁺-doped ZBLAN fiber lasers at 3 μm ," *Opt. Mater. Express* **3**, 1365-1377 (2013).
280. N. Tolstik, E. Sorokin, and I. T. Sorokina, "Graphene mode-locked Cr:ZnS laser with 41 fs pulse duration," *Opt. Express* **22**, 5564-5571 (2014).
281. W. Hanshuo, S. Jiaxin, W. Jian, X. Jiangming, X. Hu, L. Jinyong, and Z. Pu, "High-power highly stable passively Q-switched fiber laser based on monolayer graphene," *Laser Physics Letters* **15**, 035102 (2018).
282. A. G. Okhrimchuk and P. A. Obraztsov, "11-GHz waveguide Nd:YAG laser CW mode-locked with single-layer graphene," *Scientific Reports* **5**, 11172 (2015).
283. B. In Hyung, L. Hwang Woon, B. Sukang, H. Byung Hee, A. Yeong Hwan, Y. Dong-Il, and R. Fabian, "Efficient Mode-Locking of Sub-70-fs Ti:Sapphire Laser by Graphene Saturable Absorber," *Applied Physics Express* **5**, 032701 (2012).
284. I. Baylam, O. Balci, N. Kakenov, C. Kocabas, and A. Sennaroglu, "Graphene-gold supercapacitor as a voltage controlled saturable absorber for femtosecond pulse generation," *Opt. Lett.* **41**(2016).
285. Y. Fujimoto, T. Suzuki, R. A. M. Ochante, T. Hirayama, M. Murakami, H. Shiraga, M. Yoshida, O. Ishii, and M. Yamazaki, "Generation of orange pulse laser in waterproof fluoride glass fibre with graphene thin film," *Electronics Letters* **50**, 1470-1472 (2014).
286. S. Kajikawa, M. Yoshida, O. Ishii, M. Yamazaki, and Y. Fujimoto, "Visible Q-switched pulse laser oscillation in Pr-doped double-clad structured waterproof fluoride glass fiber with graphene," *Optics Communications* **424**, 13-16 (2018).
287. A. Marini, J. D. Cox, and F. J. García de Abajo, "Theory of graphene saturable absorption," *Physical Review B* **95**, 125408 (2017).
288. W. Li, J. Peng, Y. Zhong, D. Wu, H. Lin, Y. Cheng, Z. Luo, J. Weng, H. Xu, and Z. Cai, "Orange-light passively Q-switched Pr³⁺-doped all-fiber lasers with transition-metal dichalcogenide saturable absorbers," *Opt. Mater. Express* **6**, 2031-2039 (2016).
289. M. Gaponenko, P. W. Metz, A. Härkönen, A. Heuer, T. Leinonen, M. Guina, T. Südmeyer, G. Huber, and C. Kränkel, "SESAM mode-locked red praseodymium laser," *Opt. Lett.* **39**, 6939-6941 (2014).
290. Y. Cheng, J. Peng, X. Bin, H. Xu, Z. Cai, and J. Weng, "Passive Q-switching of Pr:LiYF₄ orange laser at 604nm using topological insulators Bi₂Se₃ as saturable absorber," *Optics & Laser Technology* **88**, 275-279 (2017).
291. V. G. Savitski, P. J. Schlosser, J. E. Hastie, A. B. Krysa, J. S. Roberts, M. D. Dawson, D. Burns, and S. Calvez, "Passive Mode-Locking of a Ti:Sapphire Laser by InGaP Quantum-Dot Saturable Absorber," *IEEE Photonics Technology Letters* **22**, 209-211 (2010).
292. H.-y. Lin, W.-s. Li, J.-l. Lan, X.-f. Guan, H.-y. Xu, and Z.-p. Cai, "All-fiber passively Q-switched 604 nm praseodymium laser with a Bi₂Se₃ saturable absorber," *Appl. Opt.* **56**, 802-805 (2017).
293. D. Wu, Z. Cai, Y. Zhong, J. Peng, Y. Cheng, J. Weng, Z. Luo, and H. Xu, "Compact Passive Q-Switching Pr³⁺-Doped ZBLAN Fiber Laser With Black Phosphorus-Based Saturable Absorber," *IEEE Journal of Selected Topics in Quantum Electronics* **23**, 7-12 (2017).
294. Y. Chen, G. Jiang, S. Chen, Z. Guo, X. Yu, C. Zhao, H. Zhang, Q. Bao, S. Wen, D. Tang, and D. Fan, "Mechanically exfoliated black phosphorus as a new saturable absorber for both Q-switching and Mode-locking laser operation," *Opt. Express* **23**, 12823-12833 (2015).

295. J. Chang, Z. Yang, H. Li, and M. Yang, "Passively Q-switched Nd:YVO₄/PPLN green laser with a few-layered MoS₂ saturable absorber," *Optical Review* **24**, 765-771 (2017).
296. Z. Luo, D. Wu, B. Xu, H. Xu, Z. Cai, J. Peng, J. Weng, S. Xu, C. Zhu, F. Wang, Z. Sun, and H. Zhang, "Two-dimensional material-based saturable absorbers: towards compact visible-wavelength all-fiber pulsed lasers," *Nanoscale* **8**, 1066-1072 (2016).
297. Y. Cheng, H. Yang, B. Xu, H. Xu, Z. Cai, and R. Moncorgé, "Passive Q-switching of a diode-pumped Pr:LiYF₄ visible laser using WS₂ as saturable absorber," in *Advanced Solid State Lasers*, OSA Technical Digest (online) (Optical Society of America, 2015), ATh2A.10.
298. Y. Zhang, S. Wang, H. Yu, H. Zhang, Y. Chen, L. Mei, A. Di Lieto, M. Tonelli, and J. Wang, "Atomic-layer molybdenum sulfide optical modulator for visible coherent light," *Scientific Reports* **5**, 11342 (2015).
299. K. Wu, B. Chen, X. Zhang, S. Zhang, C. Guo, C. Li, P. Xiao, J. Wang, L. Zhou, W. Zou, and J. Chen, "High-performance mode-locked and Q-switched fiber lasers based on novel 2D materials of topological insulators, transition metal dichalcogenides and black phosphorus: review and perspective (invited)," *Optics Communications* **406**, 214-229 (2018).
300. W. Fengqiu, "Two-dimensional materials for ultrafast lasers," *Chinese Physics B* **26**, 034202 (2017).
301. L. Xiaofeng, G. Qiangbing, and Q. Jianrong, "Emerging Low-Dimensional Materials for Nonlinear Optics and Ultrafast Photonics," *Advanced Materials* **29**, 1605886 (2017).
302. Y. D. Glinka, S. Babakiray, T. A. Johnson, A. D. Bristow, M. B. Holcomb, and D. Lederman, "Ultrafast carrier dynamics in thin-films of the topological insulator Bi₂Se₃," *Applied Physics Letters* **103**, 151903 (2013).
303. J. L. Wang, L. Guo, C. Ling, Y. M. Song, X. F. Xu, Z. H. Ni, and Y. F. Chen, "Carrier dynamics in femtosecond-laser-excited bismuth telluride," *Physical Review B* **93**, 155306 (2016).
304. "Observation of large nonlinear responses in a graphene-Bi₂Te₃ heterostructure at a telecommunication wavelength," *Applied Physics Letters* **108**, 221901 (2016).
305. A. Bansil, H. Lin, and T. Das, "Colloquium: Topological band theory," *Reviews of Modern Physics* **88**, 021004 (2016).
306. G. Sobon, "Mode-locking of fiber lasers using novel two-dimensional nanomaterials: graphene and topological insulators [Invited]," *Photon. Res.* **3**, A56-A63 (2015).
307. M. Hajlaoui, E. Papalazarou, J. Mauchain, G. Lantz, N. Moisan, D. Boschetto, Z. Jiang, I. Miotkowski, Y. P. Chen, A. Taleb-Ibrahimi, L. Perfetti, and M. Marsi, "Ultrafast Surface Carrier Dynamics in the Topological Insulator Bi₂Te₃," *Nano Letters* **12**, 3532-3536 (2012).
308. J. E. Moore, "The birth of topological insulators," *Nature* **464**, 194 (2010).
309. H. Zhang, C.-X. Liu, X.-L. Qi, X. Dai, Z. Fang, and S.-C. Zhang, "Topological insulators in Bi₂Se₃, Bi₂Te₃ and Sb₂Te₃ with a single Dirac cone on the surface," *Nature Physics* **5**, 438 (2009).
310. X. Wang, J. Xu, S. Gao, Y. Sun, Z. Zhu, Z. You, and C. Tu, "Dual-wavelength passively Q-switched bulk laser using MoS₂/graphene heterojunction," *Materials Research Bulletin* **89**, 63-67 (2017).
311. Z. Luo, Y. Li, M. Zhong, Y. Huang, X. Wan, J. Peng, and J. Weng, "Nonlinear optical absorption of few-layer molybdenum diselenide (MoSe₂) for passively mode-locked soliton fiber laser [Invited]," *Photon. Res.* **3**, A79-A86 (2015).
312. R. Wang, B. A. Ruzicka, N. Kumar, M. Z. Bellus, H.-Y. Chiu, and H. Zhao, "Ultrafast and spatially resolved studies of charge carriers in atomically thin molybdenum disulfide," *Physical Review B* **86**, 045406 (2012).
313. Q. H. Wang, K. Kalantar-Zadeh, A. Kis, J. N. Coleman, and M. S. Strano, "Electronics and optoelectronics of two-dimensional transition metal dichalcogenides," *Nature Nanotechnology* **7**, 699 (2012).
314. K. Wang, B. M. Szydłowska, G. Wang, X. Zhang, J. J. Wang, J. J. Magan, L. Zhang, J. N. Coleman, J. Wang, and W. J. Blau, "Ultrafast Nonlinear Excitation Dynamics of Black Phosphorus Nanosheets from Visible to Mid-Infrared," *ACS Nano* **10**, 6923-6932 (2016).

315. S. B. Lu, L. L. Miao, Z. N. Guo, X. Qi, C. J. Zhao, H. Zhang, S. C. Wen, D. Y. Tang, and D. Y. Fan, "Broadband nonlinear optical response in multi-layer black phosphorus: an emerging infrared and mid-infrared optical material," *Opt. Express* **23**, 11183-11194 (2015).
316. B. Chen, X. Zhang, K. Wu, H. Wang, J. Wang, and J. Chen, "Q-switched fiber laser based on transition metal dichalcogenides MoS₂, MoSe₂, WS₂, and WSe₂," *Opt. Express* **23**, 26723-26737 (2015).
317. J. Sotor, G. Sobon, and K. M. Abramski, "Sub-130 fs mode-locked Er-doped fiber laser based on topological insulator," *Opt. Express* **22**, 13244-13249 (2014).
318. J. Sotor, G. Sobon, M. Kowalczyk, W. Macherzynski, P. Paletko, and K. M. Abramski, "Ultrafast thulium-doped fiber laser mode locked with black phosphorus," *Opt. Lett.* **40**, 3885-3888 (2015).
319. K. Wang, J. Wang, J. Fan, M. Lotya, A. O'Neill, D. Fox, Y. Feng, X. Zhang, B. Jiang, Q. Zhao, H. Zhang, J. N. Coleman, L. Zhang, and W. J. Blau, "Ultrafast Saturable Absorption of Two-Dimensional MoS₂ Nanosheets," *ACS Nano* **7**, 9260-9267 (2013).
320. O. L. Antipov, A. A. Novikov, N. G. Zakharov, A. P. Zinoviev, H. Yagi, N. V. Sakharov, M. V. Kruglova, M. O. Marychev, O. N. Gorshkov, and A. A. Lagatskii, "Efficient 2.1- μ m lasers based on Tm³⁺:Lu₂O₃ ceramics pumped by 800-nm laser diodes," *Physica Status Solidi C: Current Topics in Solid State Physics*, Vol 10, No 6 **10**, 969-973 (2013).
321. M. J. Damzen, G. M. Thomas, and A. Minassian, "Diode-side-pumped Alexandrite slab lasers," *Opt. Express* **25**, 11622-11636 (2017).
322. S. T. Lai and M. L. Shand, "High efficiency cw laser-pumped tunable alexandrite laser," *Journal of Applied Physics* **54**, 5642-5644 (1983).
323. R. Scheps, B. M. Gately, J. F. Myers, J. S. Krasinski, and D. F. Heller, "Alexandrite laser pumped by semiconductor lasers," *Applied Physics Letters* **56**, 2288-2290 (1990).
324. F. Canbaz, N. Kakenov, C. Kocabas, U. Demirbas, and A. Sennaroglu, "Graphene mode-locked Cr:LiSAF laser at 850 nm," *Opt. Lett.* **40**, 4110-4113 (2015).
325. E. P. Ippen and C. V. Shank, "Techniques for Measurement," in *Ultrashort Light Pulses: Picosecond Techniques and Applications*, S. L. Shapiro, ed. (Springer Berlin Heidelberg, Berlin, Heidelberg, 1977), pp. 83-122.
326. D. J. Bradley and G. H. C. New, "Ultrashort pulse measurements," *Proceedings of the IEEE* **62**, 313-345 (1974).
327. R. Trebino, K. W. DeLong, D. N. Fittinghoff, J. N. Sweetser, M. A. Krumbugel, B. A. Richman, and D. J. Kane, "Measuring ultrashort laser pulses in the time-frequency domain using frequency-resolved optical gating," *Review of Scientific Instruments* **68**, 3277-3295 (1997).
328. R. Trebino and D. J. Kane, "Using phase retrieval to measure the intensity and phase of ultrashort pulses: frequency-resolved optical gating," *Journal of the Optical Society of America A* **10**, 1101-1111 (1993).
329. J.-C. M. Diels, J. J. Fontaine, I. C. McMichael, and F. Simoni, "Control and measurement of ultrashort pulse shapes (in amplitude and phase) with femtosecond accuracy," *Appl. Opt.* **24**, 1270-1282 (1985).
330. C. Iaconis and I. A. Walmsley, "Spectral phase interferometry for direct electric-field reconstruction of ultrashort optical pulses," *Opt. Lett.* **23**, 792-794 (1998).
331. R. Trebino, "FROG," in *Frequency-Resolved Optical Gating: The Measurement of Ultrashort Laser Pulses*, R. Trebino, ed. (Springer US, Boston, MA, 2000), pp. 101-115.

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PUBLICATIONS:

- C. Cihan, C. Kocabas, U. Demirbas, and A. Sennaroglu, "Graphene mode-locked femtosecond Alexandrite laser," *Opt. Lett.* 43, 3969-3972 (2018).
- C. Cihan, A. Muti, I. Baylam, A. Kocabas, U. Demirbas, and A. Sennaroglu, "70 femtosecond Kerr-lens mode-locked multipass-cavity Alexandrite laser," *Opt. Lett.* 43, 1315-1318 (2018).
- C. Cihan, E. Beyatli, F. Canbaz, L. Chen, B. Sumpf, G. Erbert, A. Leitenstorfer, F. X. Kärtner, A. Sennaroglu, and U. Demirbas, "Gain-Matched Output Couplers for Efficient Kerr-Lens Mode-Locking of Low-Cost and High-Peak Power Cr:LiSAF Lasers," *IEEE Journal of Selected Topics in Quantum Electronics* 21, 94-105 (2015).
- C. Cihan and D. Arifler, "Influence of phase function on modeled optical response of nanoparticle-labeled epithelial tissues," *Journal of Biomedical Optics* 16(8), 085002 (2011). DOI:10.1117/1.3608999

PROCEEDINGS:

- I. Baylam, A. Muti, C. Cihan, S. Ozharar, C. Kocabas, and A. Sennaroglu, "Femtosecond Pulse Generation with Graphene-Gold Supercapacitor Saturable Absorbers near 800 nm," in Lasers Congress 2016 (ASSL, LSC, LAC), OSA Technical Digest (online) (Optical Society of America, 2016), ATu3A.2.DOI: 10.1364/ASSL.2016.ATu3A.2
- C. Cihan, E. Beyatli, F. Canbaz, L. J. Chen, B. Sumpf, G. Erbert, A. Leitenstorfer, F. X. Kärtner, A. Sennaroglu, and U. Demirbas, "Gain-matched output couplers (GMOCs) for efficient and robust Kerr-lens mode-locking of Cr:LiSAF lasers," in 2014 Conference on Lasers and Electro-Optics (CLEO) - Laser Science to Photonic Applications, 2014), 1-2.
- C. Cihan and D. Arifler, "Model-based spectral analysis of photon propagation through nanoparticle-labeled epithelial tissues," in European Conferences on Biomedical Optics, (SPIE, 2011), 6.