Novel Saturable Absorber Devices Grown Using RF and Magnetron RF Sputtering

by

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B.S.E.E., University of Texas at Austin (1997)

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Abstract

Semiconductor saturable absorbers are an important technology in ultrashort pulse generation. They make ultrafast lasers self-starting, more stable, and less sensitive to cavity alignment. Epitaxially grown semiconductor saturable absorbers, although very successful, have some important disadvantages, including lattice matching requirements between the absorber and substrate and the complexity and expense of the fabrication systems.

In previous work, nonepitaxially grown semiconductor-doped silica film saturable absorbers were developed as an alternative to epitaxially grown absorbers. These absorbers consisted of InAs nanocrystallites doped into silica films and grown using rf sputtering. Rapid thermal annealing was used to control the absorption saturation dynamics. These films were fully characterized and applied to Ti:Al₂O₃ laser modelocking, producing 25 fs pulses tunable from 800 to 880 nm. The primary difficulty with these devices was their high saturation fluence, making modelocking impossible without the aid of Kerr lens modelocking (KLM).

In this work, three approaches were investigated to advance this technology. The first used a magnetron rf sputtering system to deposit GaSb-doped silica films that were expected to have a lower saturation fluence. The linear and nonlinear optical properties of these devices were fully characterized. It was found that rapid thermal annealing reduced the optical quality of these devices, making them unsuitable for laser modelocking. This was confirmed by incorporating the devices into a Ti:Al₂O₃ laser cavity, where no self-starting was observed. Using magnetron rf sputtering to deposit InAs-doped silica films should lead to improvements in device performance.

The next approach was to incorporate the InAs-doped films into a reflective geometry, with an expected decrease in the saturation fluence by a factor of four. Reflective saturable absorber devices on gold mirrors were fabricated and their linear and nonlinear optical properties were characterized. It was found that some devices were suitable for laser modelocking; however, when used in a laser cavity self-starting was not obtained. Fabry-Perot etalon effects were investigated to explain these discrepancies and it was found that this was a possible explanation for difference in the absorption saturation dynamics between devices.

Finally, InAs-doped films were deposited directly on specially designed dielectric mirrors that withstood annealing. These devices were also characterized and applied to laser modelocking. It was predicted from the linear and nonlinear optical properties and confirmed experimentally that these devices were not suitable for laser modelocking. However, devices annealed at 750 °C appear to have properties suitable for self-starting KLM and will be tested in the near future. In summary, this thesis has extended the previous work on nonepitaxially grown saturable absorbers, giving insight on important issues to consider in their design and operation.

Thesis Supervisor: James G. Fujimoto
Title: Professor of Electrical Engineering
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I will keep these acknowledgements brief, for I hope to go through the whole process again in a few years with my Ph. D. thesis.

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Chapter 1

Introduction

Ultrafast laser physics has progressed tremendously since the first demonstration of modelocking in 1964 [1]. Advances in ultrafast technology have resulted in the development of simpler, more robust femtosecond lasers producing pulses with record durations [2] [3]. In particular, significant progress has been made within the past 10 years. The use of a Ti:Al$_2$O$_3$ laser crystal [4], the Kerr lens modelocking (KLM) technique [5], and chirped mirrors for dispersion control [6] have resulted in pulse durations of 5.5 fs directly from a laser cavity [2] with no additional pulse shaping. These improvements have led to the widespread use of ultrashort pulsed lasers for many applications including optical data storage [7], THz generation and imaging [8], time-resolved spectroscopy [9] and optical coherence tomography [10].

The recent development of semiconductor saturable absorbers was also a major step in ultrashort pulse generation. These devices, known as semiconductor saturable absorber mirrors (SESAMs) [11] or saturable Bragg reflectors (SBRs) [12], typically consist of semiconductor quantum wells grown by molecular or chemical beam epitaxy and incorporated into a semiconductor mirror structure. Semiconductor saturable absorbers offer several advantages when compared to KLM. SESAMs and SBRs provide self-starting modelocking operation, and decouple the gain and modelocking mechanisms. Semiconductor saturable absorbers also simplify the cavity design and make the laser more robust and insensitive to external perturbations.

Epitaxially grown saturable absorbers, although very successful, have some disadvantages. Lattice matching imposes constraints on both the absorber and the substrate materials. The systems used for epitaxial growth are also relatively complex and expensive. Finally, some of these devices also have poor dispersion properties and narrow bandwidth of operation.
Semiconductor-doped silica thin films provide an attractive alternative to epitaxially grown saturable absorbers [13]. These devices consist of semiconductor nanocrystallites doped into a silica film and deposited on a substrate. Semiconductor-doped silica films have several advantages when compared to epitaxially grown absorbers. Rf-sputtering is a simpler, faster, and cheaper technique than epitaxial deposition. With this technique, the choice of the film and substrate materials is almost unlimited, with no lattice-matching constraints. The absorption edge can be controlled with an appropriate choice of the semiconductor dopant and nanoparticle size. The linear absorption coefficient can be controlled by varying the rf power and therefore the doping density, and the film thickness will then determine the total absorption. The semiconductor nanoparticles have a large distribution of sizes when doped into the film, resulting in a smooth absorption edge. This leads to a broad tuning range when the devices are used in a laser. Finally, unlike epitaxially grown absorbers, semiconductor-doped silica films have the potential to be deposited on double-chirped mirrors, making this technology even more powerful.

This thesis describes the further development of semiconductor-doped silica film saturable absorbers. In chapter 2, an overview of passive modelocking is presented, with emphasis on Kerr lens modelocking (KLM). Desired properties of saturable absorbers for laser modelocking are also described in detail.

Chapter 3 discusses previous work on semiconductor saturable absorbers. Epitaxially grown saturable absorbers are presented in historical order along with important results from modelocking of various solid-state laser systems. The previous development of non-epitaxially grown semiconductor saturable absorbers on which the current work is based is also discussed in detail.

Three approaches to improving semiconductor-doped silica film saturable absorbers were investigated in detail based on the conclusions from the previous work. The first approach, described in chapter 4, was to use a magnetron rf-sputtering system to make semiconductor-doped thin films for use in a transmissive geometry. This was expected to overcome some of the problems of the previous devices, particularly the high saturation fluence of the absorbers, due to the greater film purity and the increased control over deposition parameters provided by the magnetron sputtering system. When incorporated into a Ti:Al₂O₃ laser cavity, these absorbers should
make self-starting easier and possibly modelock the laser without the aid of KLM. This chapter describes the fabrication, characterization of both linear and nonlinear optical properties, and application to Ti:Al₂O₃ laser modelocking of these devices.

Chapter 5 describes the development of non-epitaxially grown saturable absorber devices for use in a reflective geometry. This would make the laser more compact, as well as allowing the saturation fluence of the devices to be controlled by placing the semiconductor-doped silica films in a particular point in the standing wave pattern of the electric field. In addition, problems previously encountered in the transmissive geometry with self-phase modulation in the sapphire substrate would be eliminated.

The first part of the chapter describes the fabrication and characterization of reflective saturable absorber devices grown on gold mirrors, all within the non-magnetron rf sputtering system. The application of these devices to Ti:Al₂O₃ laser modelocking will also be discussed. The second part of chapter 5 discusses the design and fabrication of dielectric mirror absorbers consisting of semiconductor-doped silica films grown on dielectric mirrors from Coherent, Inc. The optical properties of these devices were also comprehensively characterized and the dielectric mirror absorbers were incorporated into a Ti:Al₂O₃ laser cavity.

Chapter 6 will summarize the results of this work and suggest directions for future investigations.
Chapter 2

Passive Modelocking with Saturable Absorbers

This chapter will give a brief review of passive modelocking, focusing specifically on Kerr lens modelocking (KLM). The desired properties of a saturable absorber used in laser modelocking will also be described in detail.

2.1 Passive Modelocking

There exist many different reviews of passive modelocking [14] [15] [16] [17]; therefore, this chapter will only give a brief description. In a continuous wave (cw) laser, there are typically many different modes oscillating in the cavity. The laser is said to be modelocked when the phases of all the modes are fixed with respect to one another. This leads to a pulsed output, with time between pulses equal to the cavity round trip time; typically the process is viewed as the travel of one pulse around the laser cavity. The pulsewidth is inversely proportional to the total modelocked bandwidth.

Passive modelocking uses an intensity dependent loss or gain to favor pulsed operation over cw operation. Most methods utilize an intracavity saturable absorber, which is an element that has lower loss for higher intensities. The saturable absorber can be classified as fast or slow depending on its recovery time as compared to the pulse duration. The recovery time of a fast saturable absorber is short compared to the pulse duration. A slow saturable absorber recovers on a time scale much longer than the pulse width. Slow saturable absorber modelocking [18] was common in dye lasers, where the interplay between the saturable loss and dynamic gain saturation created a short temporal window with net gain, during which a pulse could grow. However,
dynamic gain saturation does not occur in solid-state lasers on this time scale and therefore they cannot be modelocked by this technique.

Fast saturable absorber modelocking [19] is the most common technique for modelocking solid state lasers. In addition to the saturable absorber action, pulse shaping mechanisms such as self phase modulation and group velocity dispersion influence the pulse duration and spectral bandwidth. These processes, along with gain dispersion, linear gain and loss, and self-amplitude modulation, are incorporated into the master equation for fast saturable absorber modelocking [20] [21] [22]. The master equation is then solved to obtain pulse parameters including duration, chirp, bandwidth, and stability.

2.2 Kerr Lens Modelocking

The most common technique for fast saturable absorber modelocking is Kerr lens modelocking (KLM) [5] [23]. KLM is an artificial fast saturable absorber that relies on self-focusing inside the gain crystal caused by a nonlinear index of refraction, referred to as the Kerr nonlinearity. KLM can use either a "hard" or a "soft" aperture; with a "hard" aperture, the cw component experiences greater loss than the pulsed component of radiation within the cavity, while in "soft" aperture KLM, the laser is aligned such that the mode size of the pump beam is smaller than that of the cw laser radiation. Self-focusing causes pulses to have a smaller mode size than the cw radiation within the gain medium, therefore overlapping better with the pump mode and experiencing more gain. An artificial fast saturable absorber is created with either method, since an intensity dependent gain or loss is used to select pulsed over cw operation. The response time of the nonlinearity is on the order of a few femtoseconds [24].

One disadvantage of KLM is that it is rarely self-starting. In general, starting of the modelocking is easier for gain media with lower gain cross-sections, lower saturation intensity, and shorter perturbation width [25] [26] [27]. A threshold intracavity power may also be required. For KLM, self-focusing effects are typically too weak in the cw regime to start modelocking. KLM also requires a very sensitive cavity alignment to obtain modelocking. Saturable absorbers can help overcome some of these difficulties when incorporated into a KLM laser cavity.
2.3 Saturable Absorbers for Laser Modelocking

Saturable absorbers can be used in three different regimes for laser modelocking. The first is where the saturable absorber starts modelocking for low intracavity intensites, but only weakly supports further pulse shaping. The second regime is when another mechanism starts modelocking and the saturable absorber shortens the pulse duration for higher intracavity intensities. Finally, an absorber can also be designed to perform both functions [11].

Saturable absorbers used in laser modelocking are designed to optimize several important parameters. These include linear absorption, absorption saturation intensity and fluence, absorption saturation recovery dynamics, ratio of saturable to nonsaturable losses, modulation depth, bandwidth of operation, wavelength dependence of the absorption, and damage threshold.

2.3.1 Linear Absorption and Wavelength Dependence

The saturable absorber must provide optical absorption at the lasing wavelength. The absorption must be low enough to allow the laser to reach both the lasing and modelocking thresholds. It must also be high enough to provide significantly more loss for lower intensities than higher intensities. An ideal saturable absorption would also be wavelength independent; otherwise it acts as a filter by introducing too much absorption for the laser to reach threshold at certain wavelengths. An example is the Kerr nonlinearity, an artificial fast saturable absorber which is wavelength independent in principle. However, with semiconductor saturable absorbers, the bandgap of the absorber introduces a wavelength dependence of absorption, saturation fluence, modulation depth, and nonsaturable losses. This can limit the minimum pulsewidth for sub-10 fs pulses [28].

2.3.2 Nonsaturable Losses

Ideally, the nonsaturable losses of an absorber should be made as small as possible. Nonsaturable losses make the laser less efficient, therefore requiring higher pumping power. This also makes the laser operate fewer times above threshold, increasing the tendency for instabilities such as Q-switched modelocking (to be discussed in more detail later). Nonsaturable losses are usually
caused by scattering defects in the material or free carrier absorption and can sometimes lower the
damage threshold for the device.

### 2.3.3 Saturation Fluence, Saturation Intensity, and Modulation Depth

The saturation intensity, saturation fluence, and modulation depth are three of the most
important parameters when designing a saturable absorber for laser modelocking. In the initial
stages of pulse evolution, pulses are formed by noise fluctuations in the laser. In this case, the sat-
urable absorber acts like a fast saturable absorber since the width of the fluctuations in time is
long compared to the recovery time of the absorber, and therefore the cw intensity incident on the
absorber determines the amount of absorber saturation. The saturation intensity is given by

\[
I_{sat} = \frac{h\nu}{\sigma_A T_A}
\]

where \( h\nu \) is the photon energy, \( \sigma_A \) is the absorption cross section, and \( T_A \) is the recovery time
of the absorber. The absorption coefficient saturates with the cw intensity \( I \) as

\[
\alpha = \frac{\alpha_0}{1 + \frac{I}{I_{sat}}}
\]

where \( \alpha_0 \) is the small signal absorption coefficient. As the intensity increases, the absorption
coefficient approaches zero, saturating the absorber. In general, the saturation intensity should be
high enough such that the absorber is not saturated by the cw intensity, since otherwise there
would not be enough modulation in absorption to form pulses from the noise fluctuations. Figure
2-1 shows the variation of absorption with cw intensity.
FIGURE 2-1: Nonlinear absorption change of a saturable absorber due to bleaching with the cw intensity.

For pulses comparable to or shorter than the absorber recovery time, the saturable absorber acts as a slow saturable absorber and saturates with the pulse fluence, $E_P$. The saturation behavior is then given by

$$\alpha = \alpha_0 \exp\left(-\frac{\Gamma}{\Gamma_{sat}}\right)$$

(2-3)

with $\Gamma_{sat} = \frac{h\nu}{\sigma_A}$. Figure 2-2 shows the absorption change of a saturable absorber with pulse fluence [11].

The modulation depth of a saturable absorber is the total amount of saturable losses of the absorber. This can be completely saturated by an infinitely high pulse energy, or in practice, a pulse energy much higher than the saturation fluence of the absorber. In figure 2-2, the modulation depth for a given pulse energy is the difference between the amount of absorption for low incident fluence and the amount of absorption at the pulse energy of interest. In general, the minimum pulsewidth is inversely proportional to the modulation depth for a given saturable absorber.
Minimum bounds for the saturation intensity and fluence are given by the condition for no Q-switching [11]. For the saturation intensity, the condition given by

$$\left| \frac{dA}{dI} \right| I < \frac{T_R}{\tau_2}$$

(2-4)

must be satisfied to prevent Q-switching. Here $T_R$ is the cavity round-trip time, $r$ is the pump parameter determining how many times the laser is pumped above threshold, and $\tau_2$ is the upper state lifetime of the gain medium. From figure 2-1 and equation (2-4), it is clear that Q-switching can be suppressed by using a device with a large saturation intensity (so the slope $dA/dI$ is small), a laser pumped far above threshold (to obtain a large $r$), or a very low repetition rate laser. Physically, this equation can be interpreted by examining the left and right hand sides independently. The left-hand side of (2-4) determines the amount of reduction in losses due to the bleaching of the absorber in each cavity round-trip. The intracavity intensity will increase with the loss reduction. The right-hand side gives a measure of how fast the gain saturates to compensate for the reduced cavity losses. If the gain responds too slowly, then the intracavity intensity will increase until the absorber is completely saturated and Q-switching occurs.
When the absorber recovery time is shorter than the cavity round-trip time, the condition given by (2-4) is usually fulfilled, but another condition must be satisfied to prevent Q-switching, given by

\[
\left| \frac{dA}{dT_p} \right| E_p < T_R \tau_2
\]  

(2-5)

This condition can be satisfied by choosing the pulse energy \( E_p \) very high, so that the slope of the curve in figure 2-2 is very small. Equivalently, the saturation fluence can be chosen to be very low. From figure 2-2 it can also be seen that the modulation depth increases with pulse energy before the absorber is completely bleached; therefore, operating at higher pulse energies leads to shorter pulses.

An upper limit to \( E_p \) is given by the onset of multiple pulsing. When the absorber is completely bleached, the absorption is not a strong function of pulse energy. Shorter pulses with larger bandwidth also see a reduced average gain due to the limited gain bandwidth of the laser medium. Therefore, beyond a certain pulse energy two pulses with a smaller spectrum, lower energy, and longer duration see a larger average gain, but the same amount of absorption, than a single pulse. This causes multiple pulsing to occur and sets an upper limit on pulse energies and a lower limit on the saturation fluence \([11][29]\).

In summary, there are many factors to consider when designing the saturation intensity and fluence of an absorber. The saturation intensity must be higher than the cw intracavity intensity. The saturation fluence should be low enough to avoid Q-switching instabilities and facilitate self-starting, but not so low that multiple pulsing occurs. The saturation fluence of the absorber is generally fixed by material parameters; however, the saturation intensity can be independently controlled by varying the recovery time of the absorber, if that is possible. In epitaxially grown semiconductor saturable absorbers this is done using techniques such as low temperature growth. This is probably the most common technique since it can be easily integrated with epitaxial fabrication techniques and does not cause significant amounts of material damage. Finally, the modulation depth of the absorber should be as high as possible to support short pulses.
2.3.4 Absorption Saturation Recovery Time

This parameter has an important influence on self-starting performance, modelocking build-up time, and pulse duration. In a fast saturable absorber model, the minimum pulsewidth is determined by the recovery time of the absorber. A slow saturable absorber can also be used to support soliton modelocking and generate ultrashort pulses [30].

The absorption saturation recovery time, $\tau$, should be faster than the cavity round-trip time to provide sufficient discrimination between pulsed and cw operation. This can be seen by comparing the absorption bleaching factors for cw and pulsed operation, $x_{cw}$ and $x_p$, where

$$ x_{cw} = \frac{I}{I_{sat}} \quad \text{and} \quad x_p = \frac{\Gamma}{\Gamma_{sat}}, $$

and their relationship is given by

$$ x_p = x_{cw} \frac{T_R}{\tau} \quad (2-7) $$

Therefore, for faster carrier relaxation times the absorber can provide better discrimination between the two modes of operation.

For best self-starting performance, an absorber with a long absorption saturation recovery time is desired. This can be seen by examining a simplified expression for the modelocking build-up time, a good measure of self-starting performance:

$$ T_{mlb} \approx \frac{T_R}{\left. \frac{dA}{dl} \right|_{I=0}} I \quad (2-6) $$

Better self-starting performance is given for shorter modelocking build-up times, which occur when the slope $\frac{dA}{dl}$ is large. This slope is larger for lower saturation intensities, which occur for longer carrier recovery times or larger absorption cross-sections. For example, KLM has a fast response on the order of a few femtoseconds. This makes it unsuitable for self-starting, although it results in very short pulsewidths.
Semiconductor saturable absorbers typically have a bitemporal response [31]. When electron-hole pairs are initially excited, they typically scatter out of the initial excited state and thermalize in less than one picosecond. The slower time constant is given by the time for carrier recombination and is typically tens of picoseconds. This bitemporal response is helpful in saturable absorber applications since the slow component assists self-starting and the fast component leads to shorter pulses.

Typically quantities such as saturation fluence and absorption saturation dynamics are measured using pump-probe experiments. The $(\Delta T)/T$ or $(\Delta R)/R$ signals measured in pump probe experiments are directly proportional to the quantity $(\Delta \alpha)/\alpha$, a parameter that is dependent only on the material measured. This can be shown to be inversely proportional to the saturation fluence, with the relation is given by

$$\frac{\Gamma}{\Gamma_{sat}} = \frac{\Delta \alpha}{\alpha_0}$$

(2-7)

This makes pump probe experiments very useful in determining absorption saturation dynamics. To determine the absorption saturation recovery time, an exponential or double exponential fit to the pump probe data can be used.
Chapter 3

Previous Work on Epitaxially and Non-Epitaxially Grown Semiconductor Saturable Absorbers

Semiconductor saturable absorbers are a well-developed technology for modelocking solid state lasers. They offer many advantages when compared to other passive modelocking mechanisms, including self-starting operation and increased laser stability and robustness. When compared to KLM, they have the additional advantages of decoupling of the gain and modelocking mechanisms and relaxing of the critical cavity alignment required to implement KLM.

Until recently, the majority of semiconductor saturable absorbers were epitaxially grown. These epitaxially grown devices, called semiconductor saturable absorber mirrors (SESAM) [11] or saturable Bragg reflectors (SBR)[12], have been extremely successful, either by starting and stabilizing KLM, resulting in pulse widths as short as 5.8 fs [3], or by independently modelocking a laser, enabling more simple cavity designs and more compact lasers [11]. Recently, novel non-epitaxially grown saturable absorbers have been developed using semiconductor-doped silica films deposited on sapphire substrates [13]. This chapter will describe the development of both epitaxially and non-epitaxially grown saturable absorbers in detail.

3.1 Epitaxially Grown Semiconductor Saturable Absorbers

Semiconductor saturable absorbers have previously been used to modelock diode [32] and color-center lasers [33]. However, these bulk semiconductor absorbers typically have too low a saturation intensity and damage threshold and too high insertion loss to modelock solid-state
lasers. Antiresonant Fabry-Perot saturable absorbers (A-FPSA) overcame this difficulty by inserting the saturable absorber layer in an antiresonant cavity [34].

Historically, epitaxially grown semiconductor saturable absorbers have consisted of a highly reflecting bottom mirror, a saturable absorber layer, and a top mirror of variable reflectivity depending on the device design [11]. One or more spacer layers may be included between the two mirrors, with the thickness of the absorber and spacer layers adjusted for the device to operate at antiresonance. Operation at antiresonance reduces the intensity inside the cavity, increasing the effective saturation intensity and lowering the device loss. The intensity within the antiresonant cavity is then well below the damage threshold of the device. Operation at antiresonance also leads to a negligible group velocity dispersion introduced by the Fabry-Perot cavity [34].

The reflection bandwidth and group velocity dispersion of the device are determined by either the bandwidth of the top and bottom reflectors or the free spectral range of the Fabry-Perot cavity. This is often limited by the relatively small bandwidth and poor dispersion of the bottom mirror. In general, the reflectivity of the top mirror controls most aspects of device operation since it determines the intensity entering the device, either accentuating or deemphasizing the properties of the bottom mirror. By adjusting the reflectivity of the top mirror the effective saturation intensity of the absorber layer can also be controlled. Several specific device designs will now be discussed in more detail.

### 3.1.1 High-Finesse A-FPSA

The first demonstration of laser modelocking using semiconductor saturable absorbers was achieved using an antiresonant semiconductor Fabry-Perot saturable absorber (A-FPSA) [34] [35], now referred to as the high-finesse A-FPSA. The high finesse A-FPSA surmounted the problems of high insertion loss, low damage threshold, and low saturation intensity by using the semiconductor layer as a Fabry-Perot cavity and operating at antiresonance.

The condition for antiresonance is

\[ \Phi_{rt} = 2nkda + \phi_b + \phi_t = (2m - 1)\pi \]  

(3-1)
where $\Phi_{rt}$ is the total round trip phase change of an electromagnetic wave after traversing the structure, $d_a$ is the thickness of the Fabry-Perot cavity, and $\phi_b$ and $\phi_t$ are the phase changes at the bottom and top mirrors, respectively. After one round trip the field is in antiphase and therefore interferes destructively to reduce the intensity seen by the device. The intensity inside the device is decreased by a factor of

$$\xi = \frac{1 - R_t}{[1 + \sqrt{R_t R_b \exp(-2\alpha d)}]^2} \quad (3-2)$$

for a Fabry-Perot operated at antiresonance. $R_t$ and $R_b$ are the intensity reflectivities of the top and bottom mirrors, and $\alpha$ is the field absorption coefficient of the saturable absorber. From this equation it can be seen that as described earlier, by changing the reflectivities of the mirrors, the thickness of the absorber layer, or the absorption coefficient the amount of intensity on the absorber and therefore the effective saturation fluence can be controlled [35].

The high finesse A-FPSA was fabricated using molecular beam epitaxy (MBE) by growing a bottom distributed Bragg reflector (DBR) mirror that consists of many GaAs/AlAs pairs to provide high reflectivity. However, the DBR typically has a relatively small bandwidth and poor dispersion control due to the small index difference between the semiconductors, which must be lattice matched for epitaxial growth. The saturable absorber layer was a low temperature grown InGaAs/GaAs multiple quantum well structure deposited in the same MBE run. The low temperature growth serves to reduce the carrier lifetimes in the saturable absorber. A highly reflecting TiO$_2$/SiO$_2$ dielectric mirror was finally evaporated onto the absorber layer.

Figure 3-1 (a) shows the device design. The saturation fluence of this device was measured to be 60 $\mu$J/cm$^2$. This device was first used to modelock a Nd:YLF laser, producing 3.3 ps pulses at 1.05 $\mu$m, and has also been designed and implemented at other wavelengths [36] [37] [38] [39] [40]. One problem with this device is that the reduced intensity on the absorber leads to a modulation depth of less than 1%, which limits the minimum achievable pulsewidth [28]. However, it was used to start 19 fs KLM-assisted pulses and 40 to 90 fs pulses using soliton modelocking [41].
3.1.1 AR-Coated SESAM

The opposite limit of the A-FPSA structure is to make the reflectivity of the top mirror zero by anti-reflection (AR) coating the device (figure 3-1(b)) [42]. This would increase the modulation depth, lower the saturation intensity, and improve self-starting performance, which is measured in terms of the modelocking build-up time. These devices were also fabricated using MBE; the DBR was identical to that of the high finesse A-FPSA. The absorber layer was a single GaAs quantum well to minimize insertion losses. The Fabry-Perot cavity in this device was created by the combination of the absorber layer and transparent AlAs or AlGaAs spacer layers deposited between the absorber and the DBR. The modulation depth for this device was 4.9% and the saturation fluence was 18 μJ/cm². Self-starting soliton modelocked pulses were obtained from a Ti:Al₂O₃ laser, with pulsewidths as short as 34 fs and modelocking build-up times of 3 μs [42]. Stable mode-locking was obtained over the full cavity stability range. Although the large modulation depth can support shorter pulses, the AR-coating emphasizes the properties of the bottom DBR and its small reflection bandwidth acts as a strong limitation to pulse shortening in this design.
3.1.3 Low-Finesse A-FPSA (SBR)

The low-finesse A-FPSA, also known as a saturable Bragg reflector (SBR), is an intermediate design between the AR-coated SESAM and the high-finesse A-FPSA (figure 3-1(c)) [11] [43]. The top reflector is formed by the reflection at the semiconductor-air interface of ~30%. An important advantage of this device is that the absorber layer is a single quantum well having ~10 nm thickness, which can be placed at any point in the standing wave pattern of the DBR to control the saturation intensity seen by the absorber. Placing the quantum well closer to the surface of the DBR reduces the effective saturation intensity, while positioning the absorber layer deeper in the structure increases the saturation intensity. The device can be thought of as a DBR with a weakly perturbing saturable absorber layer [43] [12]. This design requires no post-processing and can be fabricated in one MBE run. The primary disadvantage of the device is again the bandwidth of the DBR; pulses shorter than 100 fs could not be obtained from a modelocked Cr:LiSAF laser.

3.1.4 SESAM on a Silver Mirror

Better dispersion properties were obtained by replacing the bottom DBR by a silver mirror [28] [44]. This silver mirror has extremely broad bandwidth, with a reflectivity of over 98% over a range extending from 500 nm to the far-infrared. This device requires extensive processing for fabrication since semiconductors cannot be deposited by MBE on silver. The saturable absorber layer was first grown on a GaAs substrate, which was subsequently metallized with a silver mirror and bonded to an Si substrate for heat sinking. The GaAs substrate was then etched off using a selective GaAs etching technique.

A low-finesse A-FPSA design was developed with the bottom DBR replaced by a silver mirror using the techniques described above. With this design, self-starting KLM-assisted 10 fs pulses were obtained [44]. The modulation depth of this device, 1%, was not enough to support sub-10 fs pulses without the assistance of KLM. In this case, the SESAM provided a self-starting mechanism and relaxed the critical cavity alignment constraints.

The SESAM can also be designed for resonance by AR-coating the absorber device, which removes all Fabry-Perot effects; however, the coating is only correct for one wavelength, and the wavelength dependence of the reflectivity is increased. More GaAs absorber layers were added.
to obtain a device with an increased modulation depth of 6%. This device was used to generate 13 fs soliton modelocked pulses over the full cavity stability range [30] with no assistance from KLM. The modelocking build-up time was measured to be 200 μs.

Improved dispersion compensation led to self-starting KLM-assisted 6.5 fs pulses using the low-finesse A-FPSA on a silver mirror [45]. More recently, self-starting 5.8 fs pulses were obtained from a Kerr-lens modelocked Ti:Al₂O₃ laser using a broadband SESAM on a silver mirror [3]. This device has three absorbing layers made of different materials to provide absorption over a broad bandwidth, resulting in a high modulation depth of 4% over a 400 nm bandwidth and saturation fluence of 180 μJ/cm². SESAMs and SBRs have been used to modelock solid-state lasers operating at many other wavelengths, including Cr:forsterite [36], Cr:YAG [37], Nd:glass [38], Cr:LiSAF [39], and Yb:YAG [40]; Table 3-1 summarizes these results.

Finally, new technology that impacts the field of SESAM development is the design and fabrication of semiconductor double-chirped mirrors (DCM). DCMs provide a compact way to compensate dispersion over a large reflection bandwidth and in many cases can entirely eliminate the need for dispersion compensating prisms [6]. However, they are typically made using SiO₂/TiO₂ paired layers and therefore could not previously be integrated with SESAMs to compensate dispersion and provide saturable absorption in one device. An important step in that direction has been taken with the development of semiconductor DCMs [46]. This device was fabricated using metal-organic chemical vapor deposition (MOCVD) and an AR coating was deposited on the surface. It provides a significantly larger amount of dispersion per bounce on the mirror, so that a single bounce could compensate the dispersion of an entire laser cavity. This can also potentially be integrated with SESAMs to make a very powerful technology.
TABLE 3.1: Summary of important laser mode-locked results obtained using epitaxially grown semiconductor saturable absorbers.

<table>
<thead>
<tr>
<th>Laser material</th>
<th>SA type</th>
<th>SA material</th>
<th>Type of mode locking</th>
<th>Pump type</th>
<th>Pump power [W]</th>
<th>Output power [mW]</th>
<th>Minimum pulsewidth [fs]</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti:Al$_2$O$_3$ 2 mm</td>
<td>high-finesse A-FPSA</td>
<td>LT GaAs</td>
<td>SA+KLM</td>
<td>Ar ion</td>
<td>2.3 absorbed</td>
<td>300</td>
<td>19</td>
<td>[41]</td>
</tr>
<tr>
<td>Ti:Al$_2$O$_3$ 2 mm</td>
<td>AR A-FPSA</td>
<td>LT GaAs</td>
<td>SA</td>
<td>Ar ion</td>
<td>3.2 absorbed</td>
<td>120</td>
<td>34</td>
<td>[42]</td>
</tr>
<tr>
<td>Ti:Al$_2$O$_3$ 2.3 mm</td>
<td>Ag-SESAM</td>
<td>LT GaAs</td>
<td>SA+KLM</td>
<td>Ar ion</td>
<td>3.5 absorbed</td>
<td>300</td>
<td>5.8</td>
<td>[3]</td>
</tr>
<tr>
<td>Cr:forsterite 7 mm</td>
<td>SBR</td>
<td>InGaAs/InAlAs</td>
<td>SA+KLM</td>
<td>Nd:YVO$_4$</td>
<td>5.0 absorbed</td>
<td>60</td>
<td>36</td>
<td>[36]</td>
</tr>
<tr>
<td>Cr:LiSAF 2 mm</td>
<td>low-finesse A-FPSA</td>
<td>LT GaAs</td>
<td>SA</td>
<td>diode</td>
<td>0.7 absorbed</td>
<td>125</td>
<td>45</td>
<td>[39]</td>
</tr>
<tr>
<td>Cr:YAG 20 mm</td>
<td>SBR</td>
<td>InGaAs/InAlAs</td>
<td>SA</td>
<td>Nd:YVO$_4$</td>
<td>8.0</td>
<td>70</td>
<td>110</td>
<td>[37]</td>
</tr>
<tr>
<td>Nd:glass 4 mm</td>
<td>low-finesse A-FPSA</td>
<td>LT InGaAs</td>
<td>SA</td>
<td>diode</td>
<td>1.1 absorbed</td>
<td>84</td>
<td>60</td>
<td>[38]</td>
</tr>
<tr>
<td>Yb:YAG 3.5 mm</td>
<td>low-finesse A-FPSA</td>
<td>LT InGaAs</td>
<td>SA</td>
<td>Ti:Al$_2$O$_3$ at 940 nm</td>
<td>0.9</td>
<td>170</td>
<td>570</td>
<td>[40]</td>
</tr>
</tbody>
</table>
3.2 Non-Epitaxially Grown Semiconductor Saturable Absorber Films

It is clear from the previous section that epitaxially grown semiconductor saturable absorbers have been very successful in modelocking solid state lasers. However, the epitaxial growth method imposes many constraints on device design, as well as being expensive and complicated to operate. The lattice matching requirement between the absorber and substrate limits the type of materials that can be chosen for saturable absorber applications. Finally, the bottom mirror used in SESAMs imposes additional constraints on the device. In low finesse or AR-coated A-FPSA designs using a DBR as the bottom mirror, the poor reflection bandwidth and dispersion properties of the DBR limit the pulsewidth. High finesse A-FPSA designs avoid the poor DBR properties of these devices at the expense of the modulation depth of the absorber, which also limits the pulsewidth. When the DBR is replaced by a silver mirror in other designs, the extensive post-processing required to fabricate the device make it costly and complex. Therefore, an alternative to epitaxial growth was developed. The desired properties included non-epitaxial growth, relatively simple and inexpensive device fabrication, and versatility.

Semiconductor-doped silica films grown using rf sputtering are an attractive alternative to epitaxially grown semiconductor saturable absorbers [47] [48]. Rf-sputtering is a simpler, faster, and cheaper technique than epitaxial deposition. With this technique, the choice of the film and substrate materials is almost unlimited, with no lattice-matching constraints. The films used in this work consisted of semiconductor nanocrystallites in a background silica matrix. The absorption edge can be controlled with an appropriate choice of the semiconductor dopant and nanoparticle size, since the size of the particles controls the absorption edge of the device through quantum confinement. Control of the linear absorption coefficient is obtained by varying the rf power and therefore the doping density, and the film thickness will then determine the total absorption. The semiconductor nanoparticles have a large distribution of sizes when doped into the film, leading to a smooth absorption edge. This leads to a broad tuning range when the devices are used in a laser. Finally, unlike epitaxially grown absorbers, semiconductor-doped silica films have the potential to be incorporated into dielectric devices such as SiO$_2$/TiO$_2$ double-chirped mirrors, making this technology even more powerful.
3.2.1 RF Sputtering

RF sputtering is a well known and widely used technique for thin film deposition [49]. The system consists of a chamber, typically evacuated to a pressure between $10^{-6}$ and $10^{-10}$ torr, containing two electrodes. The sputtering target, containing the material to be deposited onto the substrates, is attached to the cathode, and the substrates are usually placed on the anode. The chamber is then filled with an inert gas, typically argon, and a rf voltage is applied between the electrodes. This creates a glow discharge, the positive ions of which bombard the target. The target material is then ejected by a momentum transfer process and deposited onto the substrates.

3.2.2 Device Fabrication and Characterization

InAs was chosen as the semiconductor dopant due to its bandgap in the mid-infrared range. A target was fabricated consisting of two InAs chips glued with vacuum-safe epoxy onto a 5” diameter SiO$_2$ target. The relative surface area of InAs to SiO$_2$ was $\sim$10%. The deposition rate of this target in the rf sputtering system for an rf power of 100 W was calibrated using photolithography (details of the process are in [13]) and was found to be 20 Å/min. Samples were deposited on sapphire substrates due to the high thermal conductivity of sapphire. Devices used in characterization experiments were usually about 2000 Å thick to provide a large signal for linear and non-linear optical measurements, while samples fabricated for use in laser modelocking were 300 Å thick to introduce a single pass absorption of 2% into the laser cavity. A rapid thermal anneal (RTA) treatment was performed on the devices in nitrogen for 60 seconds at temperatures ranging from 500 to 750 °C. As will be discussed later in this section, this modified the absorption saturation dynamics of the devices and provided an easy and inexpensive way of controlling these dynamics.

A comprehensive set of film characterization measurements was performed including measurements of linear absorption, femtosecond absorption saturation dynamics, nanoparticle size, film composition, element distribution within the film, and the crystalline structure of the semiconductor nanocrystallites [13] [47]. Linear absorption measurements were first performed on the semiconductor-doped films to verify that they provided optical absorption at the lasing wavelength of 800 nm. From figure 3-2, one can determine the optical absorption edge to be 1.2 μm.
The blue shift of the absorption edge from the bulk bandgap can be used to calculate the diameter of the largest InAs particles in the films, in this case 90 Å. Therefore, it was determined that this material would be suitable for Ti:Al₂O₃ laser modelocking. The smooth absorption characteristic was also expected to lead to a broad tuning range when used for laser modelocking.

X-ray photoelectron spectroscopy (XPS) measurements were performed to find the chemical composition of the semiconductor-doped silica films. From these measurements it was found that both In and As are present in the film. By comparing the XPS spectrum of this data to that of bulk In, it can be determined that the indium and arsenic exist in the form of InAs.

Transmission electron microscopy (TEM) and scanning transmission electron microscopy (STEM) measurements were taken to determine the structural composition of the InAs-doped silica films. Sample preparation for TEM measurements is typically complicated, since the specimen can only be a few hundreds of angstroms thick to transmit a large percentage of the incident electrons and therefore specimens often have to be thinned and attached to a TEM holder. An advantage of the rf sputtering technique is that films can be sputtered directly onto TEM substrates, eliminating time spent in sample preparation. It was found from TEM measurements that as expected, the films consisted of nanoparticles in a background matrix, with a large particle size distribution and maximum particle sizes of ~80 Å that agreed well with the calculated particle sizes.

![Linear absorption spectrum of a 2500 Å thick InAs-doped silica film.](FIGURE 3-2)

FIGURE 3-2: Linear absorption spectrum of a 2500 Å thick InAs-doped silica film.
The next step in film characterization was to determine the spatial distribution of elements within the film using scanning transmission electron microscopy (STEM) in conjunction with energy dispersive x-ray spectroscopy (EDX). Figure 3-3 shows the results of these measurements. From examination of these images, it can be seen that In and As are concentrated in the same region of the film, and these positions correspond to the nanocrystallites seen in the bright field (BF) image. The distribution of O and Si (not shown) was uniform, as expected. From these images, it is verified that the films consist of InAs nanocrystallites in a silica matrix.

FIGURE 3-3: Compositional maps of an InAs-doped silica film. The image sizes are 80 by 80 nm. BF: bright field image; O: oxygen; As: arsenic; In: indium.

Electron diffraction measurements were also performed to determine the crystalline structure of the nanocrystallites. The material was found to be polycrystalline, retaining the zincblende
lattice structure. The lattice constant of the structure was calculated to be 5.82 Å, comparable to the lattice constant of 6.05 Å for bulk InAs.

Femtosecond pump-probe measurements at 820 nm were performed to characterize the nonlinear optical properties of the saturable absorber films (figure 3-4). The films that underwent an RTA treatment exhibited markedly different absorption saturation dynamics than the unannealed films. The magnitude of the \( \frac{\Delta \alpha}{\alpha} \) signal, which is inversely proportional to the absorption saturation fluence of the device, increases by a factor of 2 for the annealed films, indicating a factor of 2 decrease in the saturation fluence. The saturation fluence can be calculated from these traces and is 25 mJ/cm\(^2\). This is significantly higher than the saturation fluence of typical SES-AMs, which is on the order of 50 \( \mu \)J/cm\(^2\). The saturation fluence was independent of temperature above 500 °C. The modulation depth of these devices was measured to be 0.5%.

**FIGURE 3-4:** Absorption saturation dynamics of InAs-doped silica films for different annealing temperatures. The top two traces are vertically displaced for clarity.
The absorption saturation recovery times were found to decrease with increasing annealing temperature. The mechanism of the RTA treatment and its effect on dynamics is not exactly known, but it could be attributed to traps and recombination centers created on the surface of the InAs particles during the annealing process. Initially, RTA was investigated as a way to increase the size of the quantum dots; however, it was found to be an effective way to control absorption saturation dynamics.

In general, the high absorption saturation fluences measured in these experiments can be attributed to the short exciton dephasing time $T_2$ [13]. The absorption cross section for these homogeneously broadened transitions is inversely proportional to the homogeneous linewidth, which in turn is inversely proportional to the dephasing time [16]. Therefore, fast dephasing times lead to small absorption cross sections and high saturation fluences.

Dephasing times in quantum dots are shorter for smaller dot radius, increasing temperature, higher defect density, and with excitation farther above the bandgap [13]. Smaller dot sizes lead to faster dephasing times due to scattering on the interface defects. Increasing temperature leads to more scattering with phonons. Electron-hole pairs excited higher above the bandgap have larger momentum and therefore the time between scattering events is shorter. Finally, higher defect density will also increase the amount of scattering on defects. For CdSeS dots at room temperature, dephasing times shorter than 5 fs have been reported [50].

Rf sputtering is a rough deposition method and the dots would therefore have a relatively large amount of defects. This combined with the operation at room temperature and the small nanocrystallite sizes would result in fast dephasing times and therefore a small absorption cross section, increasing the saturation fluence. This hypothesis agrees with the observed decrease in saturation fluence after annealing, where structural defects are annealed out and the dephasing times would be longer as a result. Fabricating purer films would then result in a lower saturation fluence. The use of larger quantum dots and operation closer to the bandgap should also lower the saturation fluence.
3.2.3 Laser Modelocking Experiments

Samples were fabricated as described in the previous section, with 300 Å thick InAs-doped silica films deposited on a 0° oriented sapphire substrate (to avoid birefringence effects), annealed in nitrogen at 600-750 °C, and applied to modelocking of a Ti:Al₂O₃ laser in a transmissive geometry [13] [48]. The laser cavity was a standard z-cavity with a second fold consisting of 5 cm and 7.5 cm radius of curvature (ROC) mirrors in which the absorber was placed, oriented at Brewster’s angle. The spot size on the absorber at the focus was 25 μm to increase the incident energy on the device. With this configuration, self-starting KLM was obtained with pulsewidths as short as 25 fs and bandwidth of 53 nm (figure 3-5). The pulses were not transform-limited due to the excess self-phase modulation in the sapphire substrates. The wavelength could be tuned from 800 to 880 nm while maintaining self-starting operation.

This is believed to be the first demonstration of laser modelocking using non-epitaxially grown saturable absorbers. This promises to be a powerful and versatile technology; however, there are some potential improvements to this technique. The high saturation fluence and low modulation depth limits pulsewidth and does not enable saturable absorber modelocking without KLM. Also, when used in this transmissive geometry, the self-phase modulation in the sapphire substrate limits further pulse shortening. These issues will be addressed in the following chapters.
FIGURE 3-5: (a) Spectrum and (b) autocorrelation of self-starting modelocked pulses from KLM Ti:Al2O3 laser incorporating an InAs-doped silica film saturable absorber.
Chapter 4

Transmissive Saturable Absorber Devices Using Magnetron-Sputtered Semiconductor-Doped Silica Films

Magnetron rf sputtering offers many advantages over conventional rf sputtering that should improve the performance of semiconductor-doped silica film saturable absorbers. Deposition rates can be an order of magnitude higher with the use of a magnetron rf sputtering system. In addition, the magnets confine the plasma around the target, eliminating contact of the substrates with the plasma. This leads to more precise control of the substrate temperature with a substrate heater, also available in the magnetron system, which can increase quantum dot sizes[51] and reduce the saturation fluence as discussed in previous work [13].

4.1 Sample Fabrication

4.1.1 Description of Magnetron RF Sputtering System

Magnetron rf sputtering differs from conventional rf sputtering by the addition of magnets which confine the plasma to the vicinity of the target [52]. This results in increased deposition rates, often an order of magnitude higher than a system without a magnetron. The substrates are also isolated from the plasma discharge.

Many magnetron sputtering systems also allow control of the substrate temperature with a heating lamp installed in the vacuum chamber. In previous work [51] [53], it has been shown that nanocrystallite size increases with increasing substrate temperature, which will red shift the
absorption edge. In the earlier work on rf sputtering of semiconductor-doped silica films [13], no variance of nanocrystallite size with substrate temperature was observed. This could be due to the lack of a magnetron in the system, which resulted in lower deposition rates and also allowed the plasma to come into contact with the substrates. The plasma bombardment of the substrates was found to increase the temperature in the chamber to 70 °C, making it difficult to precisely control the substrate temperature. In a magnetron system, it should be possible to carefully control the substrate temperature and therefore the nanocrystallite sizes.

The magnetron sputtering system used in this work was located at Boston University and made by Denton Vacuum. The rf power supply was separate and made by Manitou, operating at a frequency of 13.56 MHz. This system has 3 targets, one of which was used in this work. A heating lamp was installed in the vacuum chamber to control the substrate temperature. This system also had a loadlock, which enabled the user to move substrates in and out of the chamber without pumping down the entire vacuum chamber. This made it possible to do several sputtering runs in one day, as the time to pump the loadlock down was approximately 30 minutes. Typical vacuum pressures when the system was pumped down were 5·10⁻⁷ Torr. A final advantage of this system was ability to rotate the substrate platform during deposition, leading to uniformity of film thickness between samples made in the same run.

4.1.2 Selection of Semiconductor Dopants

The choice of semiconductor dopant is governed by two factors: whether the film will have the desired saturable absorber properties and whether the semiconductor will contaminate the sputtering system. As previously discussed, a necessary characteristic of the semiconductor doped films is optical absorption at the laser wavelength. The quantum confinement effect must also be considered when choosing the semiconductor dopant, since the semiconductor bandedge in the films will be blue shifted from the bulk bandgap. For the near IR laser wavelengths considered in this work, suitable materials will probably have a bulk bandgap in the mid-IR. Vacuum chamber contamination in the sputtering system is another issue to consider. This can be avoided by choosing semiconductors that do not contain lead and sulfur.
After considering these factors, GaSb, with a bulk bandgap of 1.9 \( \mu m \), was chosen as the semiconductor dopant for our first target in the magnetron sputtering system. Semiconductors such as CdSe, with a bulk bandgap of 718 nm, would be blue shifted by quantum confinement and therefore have no appreciable absorption at 800 nm. In comparison, a semiconductor such as InSb has a bulk bandgap of 6.9 \( \mu m \), which would probably have too much absorption at 800 nm after being blueshifted. The energy shift required in GaSb to have appreciable absorption at 800 nm is significantly less than the shift required from the bulk InAs bandgap of 3.5 \( \mu m \). This should lead to larger quantum dot sizes and therefore reduce the saturation fluence by the arguments in section 3.2.2. The magnetron sputtering process would be expected to produce purer films as well, further reducing the saturation fluence.

4.1.3 Target Fabrication

The target was circular and consisted of a 3 inch diameter SiO\(_2\) disk with two semiconductor chips attached to it. The SiO\(_2\) disk was attached to a copper backing plate, and then the semiconductor chips were glued on using vacuum safe epoxy. The epoxy was able to withstand temperatures up to 100°C without loss of vacuum pressure. The semiconductor chips were cut so that the relative surface area of semiconductor on the target was \( \sim 10\% \). A diagram of the GaSb+SiO\(_2\) target is shown in Figure 4-1.

**FIGURE 4-1:** Diagram of the GaSb+SiO\(_2\) target used in the magnetron system.
4.2 Linear Transmission

The initial goal of film deposition experiments was to study the dependence of the absorption edge on deposition parameters including rf power, substrate temperature, and film thickness. Many deposition runs were performed while independently varying one or more of these parameters. Linear transmission spectra were obtained using a Cary 5E spectrophotometer. From these measurements the variance of the absorption edge with the deposition parameters could be determined.

4.2.1 Absorption Edge Variance with RF Power

There has been very little work reported in the previous literature on variations of quantum dot size with rf power [51] [53]. Several runs were performed while varying the rf power. In all runs done with the magnetron rf sputtering system, sapphire substrates with 1/2" diameter and 1/8" thickness were used. During these runs, the system pressure was kept constant at $5.2 \times 10^{-3}$ Torr and the substrate heater remained turned off, leaving the vacuum chamber at a temperature of 22 °C. Linear transmission spectra were measured after deposition. The results of these experiments are shown in figure 4-2.

![FIGURE 4-2: Transmission as a function of wavelength for different rf powers.](image-url)
From this figure, it can be seen that the linear transmission characteristics change considerably as the rf power is varied; however, some portion of the ripples in the spectra can be attributed to interference. At this point the deposition rates for different powers had not been calibrated, and therefore the thickness of the deposited films was unknown. Interference effects would become more noticeable as the film thickness increased. A better indicator of absorption edge variance with power would be to keep the film thickness constant while varying the rf power, such that interference effects should be the same for all samples.

To this end, the deposition rates were calibrated using photolithography for rf powers of 50 W, 75 W, and 150 W. The deposition rates are shown below in figure 4-3 and it can be seen that the deposition rate scales linearly with the rf power. A linear fit was applied to the data, yielding the equation shown in the figure.

![Calibrated Deposition Rates vs. Power](image)

**FIGURE 4-3:** Calibrated deposition rates vs. rf power.

Previous data on sputtering of GaSb+SiO₂ films in a non-magnetron system calibrated the deposition rate to be 27 Å/min [54]. This data shows that the deposition rates for GaSb+SiO₂ films sputtered in a magnetron system increased by a factor of 1.6 as compared to the non-magnetron system. Films were deposited with a thickness of approximately 1935 Å at different rf pow-
ers. Linear transmission spectra were taken using the Cary 5E spectrophotometer. The results of these experiments are shown in figure 4-4.

![Transmission spectra](image)

**FIGURE 4-4:** Linear transmission as a function of wavelength for different rf powers while keeping film thickness approximately constant.

From this figure, it can be seen that the absorption edge does not vary with rf power. The small differences in magnitude of transmission between the curves can be attributed to slight variations in film thickness. These experiments eliminated rf power as a deposition parameter to control quantum dot size.

### 4.2.2 Absorption Edge Variance with Film Thickness

Preliminary studies indicated that the absorption edge may vary with film thickness, an effect which had not been observed in previous studies [51] [53]. This would be detrimental to film characterization studies, since samples used in pump-probe and linear transmission measurements are typically on the order of 200 nm thick to obtain a large signal and minimize noise. In comparison, samples used in the laser are typically ~300 Å thick to make the single pass absorption around 2%. A change in the properties of the sample with thickness would make characterization of samples to be used for laser modelocking very difficult.
Several runs were performed while keeping the rf power and substrate temperature constant and varying the deposition time. The results of linear transmission spectra taken on these samples are shown in figure 4-5.

![Linear transmission versus wavelength, for different film thicknesses.](image)

**FIGURE 4-5:** Linear transmission versus wavelength, for different film thicknesses.

As in the previous experiments on absorption edge variance with rf power, interference effects made it hard to clearly identify any absorption edge dependence on thickness. The absorption coefficient $\alpha(\lambda)$ is given by

$$\alpha(\lambda) = \left(-\frac{1}{d}\right) \ln\left(\frac{I}{I_0}\right)$$

(4-1)

A plot was made to see how the total $\alpha d$ varies (figure 4-6 (a)). Interference effects were negligible in this plot, but the effects of the different film thicknesses were still apparent. By plotting $\alpha(\lambda)$ against wavelength for different film thicknesses, the dependence of the absorption coefficient on film thickness can be determined and errors in the film thickness are less important. This dependence is shown in figure 4-6 (b).

The shape of these curves is very similar, with most plots having an absorption edge around 1 $\mu$m, and the magnitudes of the curves are also comparable. This suggests that the thickness of
the films is not a strong factor in changing the absorption edge and therefore parameters obtained from characterization of thicker films such as absorption edge, saturation fluence, and absorption saturation dynamics should be applicable to thinner films.

**FIGURE 4-6:** (a) Plot of the the absorption coefficient times film thickness versus wavelength for different film thicknesses. (b) Plot of the absorption coefficient versus wavelength for different film thicknesses. The samples with thicknesses of 32.3 nm and 64.5 nm were normalized for reflection to minimize the error due to their relatively small absorptions.
4.2.3 Annealing of Magnetron Sputtered GaSb+SiO₂ Films

Rapid thermal annealing of the GaSb-doped silica films was expected to modify the absorption saturation dynamics in a similar manner as in the InAs-doped films [13]. This would make the films more suitable for saturable absorber applications by reducing the saturation fluence and decreasing the absorption saturation recovery times. The magnetron sputtered films were annealed in nitrogen at temperatures ranging from 400 to 700 °C for 60 seconds using an AG Associates Heatpulse 210 RTA system. Visual inspection showed that many of the films appeared to lose optical quality after the annealing process; the optical absorption of the films also decreased due to diffusion and oxidation, as previously observed. Linear transmission spectra were measured on the annealed films (figure 4-7).

Figure 4-7 shows the transmission spectrum of a representative GaSb-doped silica film on a sapphire substrate annealed at 500 °C for 60 seconds. It is clear that the spectrum is slightly blue-shifted and the magnitude of transmission is somewhat greater for the annealed sample; this corresponds well with results from previous work [13]. Most samples exhibited this kind of change after annealing. The spectrophotometer measurement used a 3 mm aperture, which averaged out the effects of the film damage after the RTA process. Overall, the optical quality of many samples was reduced after the RTA process, although this change was not usually
observable in linear transmission measurements and a clear dependence on rf power, annealing
temperature, or film thickness could not be identified. This contrasts with the data on non-magne-
tron sputtered InAs-doped silica films, where the RTA process had no effect on optical quality.

4.3 Nonlinear Optical Properties

Femtosecond pump probe experiments were performed on the GaSb-doped silica films, both
before and after rapid thermal annealing, to characterize their absorption saturation dynamics, for
the first time to my knowledge. As discussed in chapter 2, the magnitude of the pump probe sig-
nal is inversely proportional to the saturation fluence and an exponential curve fit to the data is a
measure of the relaxation dynamics.

The system is shown in figure 4-8 and the samples are measured in transmission. The pulses
are generated by a home-built Kerr lens modelocked Ti:Al₂O₃ laser pumped by an argon laser.
When entering the system, a pulse is split up into two pulses, the pump and the probe, which in
these experiments had approximately equal power. A variable time delay is introduced between
the pulses by changing the path length of the pump beam using a translation stage. The two
pulses are then focused onto the same spot on the sample. The pump pulses excites electron-hole
pairs within the semiconductor, changing the sample absorption by band filling. The pump-
induced change in the sample transmission or reflection is detected by the probe pulse. The pump
beam is chopped and the probe is detected by a lock-in amplifier to improve detection sensitivity.
The absorption saturation dynamics in the sample can be mapped as a function of time by varying
the delay between the pump and the probe.

The laser wavelength was 820 nm for the measurements on the unannealed samples and 800
nm for the measurements on the annealed samples. The pump and probe were cross-polarized to
avoid interference artifacts. Pulses were generated with 30 fs duration at 80 MHz repetition rate.
The pump power was 100 mW and the spot size on the sample was 30 μm. Measurements were
performed on GaSb-doped silica films grown with different rf powers and film thicknesses.
**FIGURE 4-8**: Femtosecond pump probe system used for measuring absorption saturation dynamics.

### 4.3.1 Measurements on GaSb-doped silica films before annealing

Initial pump-probe measurements on GaSb-doped silica films before annealing indicated that the \( \frac{\Delta \alpha}{\alpha} \) signal increased in magnitude by as much as a factor of 1.3 when compared to the InAs-doped silica films (figure 4-9). The relaxation times were comparable in both cases, with relatively slow time constants of hundreds of femtoseconds.

**FIGURE 4-9**: Comparison of magnetron and non-magnetron sputtered films.
A point to note is that the baseline for the GaSb+SiO$_2$ films is much larger than that for the InAs+SiO$_2$ films, possibly indicating long lived thermal effects in GaSb. In general, the baselines in pump probe measurements are due to scattering of the pump into the detector and are sensitive to system alignment. In this experiment, the samples were measured with the same system alignment, minimizing the role of pump scattering when comparing the baselines of the two samples. The thickness of the sapphire substrate was also found not to affect the magnitude of the baseline.

Pump probe measurements were also performed on samples grown with different rf powers. The magnitude of the signals did not vary strongly with rf power, and the absorption saturation recovery times were comparable for all samples.

The wavelength dependence of the absorption saturation dynamics was also characterized by tuning the laser over a range from 793 to 826 nm. The results are shown in figure 4-10. From these measurements it can be seen that there is no dependence on wavelength in this tuning range, which is also the wavelength range over which samples would be used in laser modelocking.

![Graph showing the wavelength dependence of absorption saturation dynamics.](image)

**FIGURE 4-10:** Wavelength dependence of absorption saturation dynamics of an unannealed GaSb-doped film. The film was grown at 150 W for 30 minutes.

4.3.2 Measurements on GaSb-doped silica films after annealing

45
As previously described, the optical quality of many of the GaSb-doped silica films decreased considerably after rapid thermal annealing. This made it harder to obtain pump-probe signals from the samples due to the large amount of scattering. Many of the samples were too damaged to find a focus, and clear pump probe signals could only be obtained from a few samples (figure 4-11). Before annealing it was possible to obtain a signal from all samples tested.

**FIGURE 4-11:** GaSb-doped silica films both before and after rapid thermal annealing. (a) Film grown at 100 W for 60 min and annealed at 500 °C for 60 s. (b) Film grown at 150 W for 30 min and annealed at 400 °C for 60 s. The spike in the signal is due to noise.
In figure 4-11 (a), it can be seen that the annealing process had almost no effect on the dynamics of this GaSb-doped film, with the signal magnitude and absorption saturation recovery times almost unchanged. This contrasts strongly with the effects of the rapid thermal anneal on the InAs-doped films, where the $(\Delta\alpha)/\alpha$ signal increased by a factor of 2 and the recovery times significantly decreased. This would be expected to have a strong effect on laser modelocking with magnetron sputtered GaSb films, since unannealed InAs samples did not self-start modelocking in the previous work [13].

Figure 4-11 (b) shows a representative film that was damaged by the RTA treatment and therefore had a very small pump-probe signal. Clearly the rapid thermal annealing process, which improved the absorption saturation dynamics for laser modelocking in InAs-doped silica films, has a detrimental effect on GaSb-doped silica films, either degrading the optical quality to a point where a pump probe signal cannot be obtained or leaving the absorption saturation dynamics unaffected. Therefore, these samples would not be expected to successfully self-start Ti:Al$_2$O$_3$ laser modelocking.

### 4.3.3 Coherent Phonon Oscillations

The effects of varying the substrate temperature during the film deposition process were only looked at in the early stages of this work. Initial linear transmission and pump probe measurements before the annealing showed no significant difference from measurements on other samples. However, after the RTA treatment, pump probe traces were taken again. In these measurements, large oscillations were observed in the pump probe signal (figure 4-12), with a frequency of 5 THz. This frequency is of the same order of magnitude as phonon frequencies in quantum dots measured in other materials systems [55] [56] and also similar to those of bulk GaSb [57], indicating that these may be coherent phonon oscillations. The 5 THz oscillation frequency falls within the gap between optical and acoustic phonon frequencies for bulk GaSb.

A simple model can be applied to calculate the lowest order acoustic phonon mode by viewing the nanocrystallites as homogeneous elastic spheres and the acoustic phonon vibrations as eigenmodes of the sphere in a classical approach [58]. An expression for the lowest order acous-
tic mode frequency is given by $\nu = \frac{c_L \chi}{R}$, with $R$ the radius of the nanocrystallite and $\chi$ defined by

$$\tan \chi = \frac{\chi}{1 - \frac{c_L}{4c_T} \chi^2}$$  \hspace{1cm} (4-2)

where $c_L$ and $c_T$ are the longitudinal and transverse wave velocities in the material, given by $c_L = \sqrt{\frac{C_{11}}{\rho}}$ and $c_T = \sqrt{\frac{C_{44}}{\rho}}$, with $\rho$ being the density of the material and $C_{11}$ and $C_{44}$ the elastic constants of the material. This equation can be numerically solved, using the values of these constants for GaSb [57] to obtain a value of $\chi = 2.12$.

**FIGURE 4-12:** Observation of oscillations in pump probe signal for sample grown with substrate heating of 100 °C and post-annealed at 500 °C.

The radius of the GaSb nanocrystallites is unknown at this point; however, based on the previous work on InAs nanocrystallites and the shift of the absorption edge from bulk GaSb, a value of 100 Å was assumed for the nanocrystallite radius as a first estimate. The frequency of the lowest order acoustic mode was calculated to be 0.8 THz, well below the observed 5 THz oscillation frequency. Possible sources of error in the calculation include the approximation that the spheres are in vacuum, instead of being surrounded by a silica matrix, and also the fact that the approximation can fail for particles consisting only of a few unit cells. However, it still seems likely that
the observed oscillations are not due to coherent acoustic phonons. Further studies of these effects will be performed in the near future.

4.4 Laser Modelocking Experiments

As previously discussed, the effect of annealing on the GaSb-doped silica films was to reduce optical quality and at best leave optical performance unaffected, often degrading it significantly. Therefore, both annealed and unannealed films would not be expected to start laser modelocking when inserted into the laser cavity.

Samples were fabricated for use in the Ti:Al₂O₃ laser cavity in a transmissive geometry. The samples were magnetron rf sputtered at 75 W rf power, grown on 3 mm thick sapphire substrates, and had thicknesses varying from 400 Å to 165 Å to provide different values of linear absorption. The substrates were 0°-oriented to avoid birefringence effects. The films were annealed at temperatures of 750 and 600 °C in nitrogen for 60 seconds. Single pass transmission was measured over the laser tuning range and was found to vary between 1% and 15% depending on film thickness.

The laser cavity is shown in figure 4-13 and is the same as that used in reference [13]. It is a standard z-cavity with 10 cm radius of curvature folding mirrors (CM1) and a second fold consisting of 5 cm and 7.5 cm focusing mirrors (CM3 and CM2, respectively). The separation between the mirrors in the second fold was approximately 10 cm in order to make its ABCD matrix unity. The saturable absorber film was positioned at Brewster's angle in the focus of the second fold; the incidence angle on CM3 was set to 13° to compensate the astigmatism introduced by the sample. The spot size of the laser mode on the absorber was approximately 25 μm. The Ti:Al₂O₃ crystal was 4.5 mm long and absorbed 80% of the incident pump radiation. Pump powers were between 5.5 and 6.5 W. Dispersion compensation was provided by two fused silica prisms separated by 76 cm.

The laser was first optimized for KLM using a blank sapphire substrate positioned at the focus of the second fold. The output power was 100 mW from a 2% output coupler with 6.5 W
pump power incident and the bandwidth of the modelocked pulses was 27 nm. When the sapphire blank was replaced by a GaSb-doped saturable absorber device, the laser did not self-start, even when the absorber was moved into the focus of the second fold. KLM could be obtained on all absorbers when laser parameters including curved mirror separation and output coupling were varied, but overall the modelocking performance was not improved. This agreed with the results of pump probe experiments and also with results reported in the work on InAs-doped silica films [13]. In order to improve the performance of GaSb-doped silica films and make them suitable for laser modelocking, an alternative to rapid thermal annealing as a way of tailoring the absorption saturation dynamics would have to be developed. Another possibility would be to use an InAs+SiO$_2$ target in the magnetron sputtering system, since it responds well to rapid thermal annealing. The control over substrate temperature and the increased deposition rates should enable the development of saturable absorber films with reduced saturation fluence, making the

**FIGURE 4-13:** Laser cavity used in experiments on modelocking with GaSb-doped silica film saturable absorbers in a transmissive geometry. CM1: 10 cm ROC dielectric mirror; CM2: 7.5 cm ROC dielectric mirror; CM3: 5 cm ROC dielectric mirror; SA: saturable absorber; OC: output coupler; P: prisms.
devices more broadly applicable to laser modelocking. Experiments exploring these possibilities are currently in progress.
The use of semiconductor-doped silica thin films in a reflective geometry would eliminate the problems with excessive self-phase modulation in the sapphire substrate that were encountered when using the transmissive geometry. This would also simplify the laser cavity design. If properly designed, reflective absorber devices could also have an increased nonlinearity and therefore a lower saturation fluence, similar to the previous work with SBRs and SESAMs.

5.1 Reflective Saturable Absorber Devices on a Gold Mirror

As previously discussed, one of the major advantages of developing saturable absorber devices using rf sputtering is the ability to deposit saturable absorber films on almost any substrate, without the lattice matching constraints encountered in work with quantum well saturable absorbers \[11\] [12]. This technology enables deposition on substrates unsuitable for epitaxial deposition, including metal and dielectric mirrors. The initial goal was to develop a simple reflective device that could be grown completely within the non-magnetron rf sputtering system at Lincoln Laboratories used in the previous work [13] and described in section 3.2.1.

5.1.1 Design and Fabrication of Gold Reflective Absorber Devices

The trial design for a gold reflective absorber consisted of a InAs doped silica film deposited directly on a gold mirror, all grown in the rf sputtering system at Lincoln Laboratories. This sys-
tem was an MRC 8800 rf sputtering system and details of operation, along with a description of the film deposition procedure, can be found in [13]. Gold was chosen due to its broad reflectivity bandwidth, with a reflectivity greater than 97% over a range from 700 nm to the mid-IR, and also because of its ready availability in the sputtering system.

A 400 Å thick titanium layer was initially deposited on top of sapphire and fused silica substrates to provide good adhesion between gold and the substrates. A 1600 Å gold film was deposited next, followed by a 300 Å thick InAs doped silica film. The thickness of the gold layer was chosen to be several times the skin depth of gold at 800 nm, equal to ~300 Å. The thickness of the InAs doped film was designed to give 2% absorption at 800 nm. The devices were subsequently annealed in nitrogen for 60 s at temperatures ranging from 500 to 700 °C to control the absorption saturation dynamics, as in the previous work. The films adhered well to the substrates and had good optical properties.

However, these devices are not suitable for use as saturable absorbers for modelocked lasers. A standing wave is formed in the laser cavity by the incident and reflected waves when an electromagnetic wave is reflected from a mirror. A node in the standing wave pattern exists at the surface of a gold mirror, effectively increasing the saturation fluence of a semiconductor-doped silica film deposited directly on the surface. Since the saturation fluence of the films measured in pump-probe experiments is already high, this design would not be optimal for laser modelocking applications. A better design would place the film at the peak of the standing wave, increasing the intensity on the film and reducing the effective saturation fluence.

Placement of the film at the peak of the standing wave would actually reduce the effective saturation fluence by a factor of 4 as compared to the transmissive geometry, making modelocking easier to start. This can be seen by considering the electric field near the mirror with incident and reflected waves having approximately equal amplitudes:

\[ E = E_0 e^{-ikz} - E_0 e^{ikz} = -2iE_0 \sin kz \]

where the reflection coefficient is -1 at the surface of the metal mirror. The intensity of the standing wave is given by

\[ I = |E|^2 = 4E_0^2 \sin^2 kz \]
\[ I = 4I_0 (\sin kz)^2 \]

which is a factor of 4 higher at the peak than the intensity \( I_0 \) on the film in the transmissive geometry. The linear absorption is only increased by a factor of 2, since the absorber is double-passed in the transmissive geometry.

To place a semiconductor-doped film at the standing wave peak, transparent quarter wave spacer layers can be used (figure 5-1). The total thickness of the structure on top of the mirror should be \( \left( \frac{\lambda}{2} \right) m \), where \( m \) is an integer, to keep the reflectivity of the total structure high. The wave reflected from the surface of the device will have a phase of \( \pi \) and the wave reflected from the gold mirror will acquire a phase of \( \pi + 2m\pi \), interfering constructively with the first wave.

![Design of a reflective saturable absorber device with sapphire spacer layers on a gold mirror.](image_url)

**FIGURE 5-1:** Design of a reflective saturable absorber device with sapphire spacer layers on a gold mirror.

The first design for this device used silica spacer layers. The deposition rate for silica was calibrated and found to be 17 Å/min. The device was fabricated by depositing titanium and gold layers as before on sapphire, fused silica, and BK-7 substrates. Deposition continued with 124 nm of SiO\(_2\), 30 nm of InAs+SiO\(_2\), and 124 nm of SiO\(_2\) to make the total optical thickness of the structure 400 nm. The total time to deposit the structure was about 3.5 hours.
The devices were annealed in nitrogen for 60 s at temperatures between 500 and 700 °C to control the absorption saturation dynamics of the saturable absorber films. After annealing, all of the films were found to be cracked and the optical quality was greatly reduced, making them unsuitable for laser modelocking applications. An image of a 640 by 480 μm region of the annealed films was acquired using a microscope and is shown in figure 5-2.

**FIGURE 5-2:** Surface of a reflective saturable absorber device with silica spacer layers after rapid thermal annealing at 500 °C for 60 seconds. The image size is 640 by 480 μm.

Interference fringes can be observed near the edges of the darker regions, which suggests that the distance between the reflective surfaces was around several wavelengths of light in the visible. Since the deposited film was only 400 nm thick, this indicates that the film was elevated above the gold reflector, probably by a few microns at the center where there are no fringes and
less at the edges. Profilometry measurements were performed on the films, which indicated that the islands on the image were elevated by approximately 3 μm above the substrate.

A possible explanation for the film damage results from the difference in the thermal expansion coefficients between gold and silica. Gold has an expansion coefficient of 14.2·10⁻⁶, while silica has an expansion coefficient of 0.51·10⁻⁶ K⁻¹ [59]. When the film is annealed, the gold layer expands more than the silica film; the silica film cannot stand the increased strain and breaks up into the observed SiO₂ islands on top of gold. After the sample cools down, the gold will contract more than the silica, causing the silica to form dome-shaped structures.

A more suitable material for the transparent spacer layers is sapphire, with a thermal expansion coefficient of ~7·10⁻⁶ K⁻¹ that is closer to the expansion coefficient of gold. The SiO₂ target was replaced by a sapphire target in the sputtering system and the deposition rate was calibrated to be 9 Å/min. Reflective saturable absorber mirror structures were fabricated, with the silica layers in the previous design replaced by sapphire. Samples were first annealed before depositing the second sapphire spacer layer and examined for optical quality. After RTA in nitrogen at temperatures ranging from 500 to 700 °C, the structures still had high optical quality and could be used in laser modelocking experiments. Samples from this run were also annealed after the second sapphire spacer layer was deposited and found to preserve high optical quality.

5.1.2 Linear Reflectivity Measurements

Preliminary experiments indicated that the samples had high linear absorption, possibly too high for Kerr lens modelocked laser operation. The linear reflection spectrum of a reflective gold saturable absorber film with a 400 Å InAs-doped film deposited on a 1” sapphire substrate was measured using the Cary 5E spectrophotometer with a reflective attachment and is shown in figure 5-3. The reflective attachment required samples to be at least 1” in diameter; therefore, many samples that were deposited on 0.5” sapphire substrates could not be measured.
FIGURE 5-3: Reflectivity of a gold saturable absorber device. The film thickness was 400 Å and the device was annealed at 600 °C prior to deposition of the second sapphire spacer layer.

The absorption of this sample was 20% at 800 nm when assuming no transmission through the gold mirror and sapphire substrate. This is higher than the expected value of approximately 12%. The peaks and valleys in the spectrum at 620 nm and 450 nm are due to interference effects. The absorption edge was at ~1200 nm as before, indicating that the sapphire spacer layers and annealing process have no strong effect on the quantum dot size. Samples were also fabricated with 200 Å thick InAs-doped silica films replacing the 400 Å films in the previous design and annealed at temperatures ranging from 500 to 700 °C after the whole structure was deposited. These structures appeared to have high optical quality that was unaffected by the rapid thermal anneal. These devices also could not be measured in the spectrophotometer since they were grown on 0.5” substrates.

5.1.3 Pump Probe Experiments and Results

Pump probe experiments were performed to measure the absorption saturation dynamics of the reflective gold absorber devices. The system was aligned to measure signals in reflection (figure 4-8) by placing a silver mirror at the focus of the 5 cm lens and realigning the 50.8 mm lens, the aperture, and the detector to obtain the maximum signal. Measurements were performed on
all reflective devices; however, with some samples the signal was too small to detect with the sen-
sitivity of this system due to the small sample absorptions. Some representative results of these
pump probe experiments are shown in figure 5-4.

Several interesting points can be noted about the nonlinear optical properties of the reflect-
tive gold absorber devices. Absorption saturation recovery dynamics were essentially equivalent
for all samples, with time constants of hundreds of femtoseconds. This corresponds well with
previous data taken on transmissive samples, indicating that the annealing process affected the
absorption saturation dynamics of the reflective gold absorbers in the same manner.

The magnitude of the \( \frac{\Delta R}{R} \) signals, inversely proportional to the saturation fluence,
was also similar for all samples but one (figure 5-4 (a). The samples annealed before the second
sapphire spacer layer was deposited (figure 5-4 (b)) exhibited comparable signal magnitudes to
the samples annealed after the whole structure was grown (figure 5-4 (c)). This implies that the
annealing process affects dynamics in the same way regardless of the point in deposition at which
the film is annealed. Film thickness did not have a significant effect on dynamics; samples with
both 200 Å and 400 Å InAs-doped silica films had similar dynamics, although the 200 Å films
had smaller signals as expected since the films are thinner.

Although the measured absorption saturation recovery times were comparable to previous
results with transmissive samples [13], the magnitude of most signals was significantly smaller
than expected. Typical peak values for \( \frac{\Delta T}{T} \) with InAs-doped films deposited directly on
sapphire substrates were \(-0.75 \cdot 10^{-3}\) for unannealed samples and \(1.5 \cdot 10^{-3}\) for annealed films. Most
annealed reflective gold absorbers yielded \( \frac{\Delta R}{R} \) signals with peak values of \(1 \cdot 10^{-3}\), implying
a saturation fluence similar to that of the unannealed transmissive films and probably not large
enough to start modelocking. This differs drastically from the expected factor of 4 decrease in
saturation fluence.
annealed at 600 C
obtained best KLM-30 mW, 46 nm
exponential time constant-430 fs

annealed at 600 C with second sapphire layer
obtained KLM, 90 mW, 42 nm
exponential time constant-433 fs

annealed at 500 C
obtained KLM, 50 mW, 46 nm
exponential time constant-307 fs

FIGURE 5-4: Reflective pump-probe traces of gold absorber devices. (a) 400 Å film, annealed at 600 °C without second sapphire layer. (b) 400 Å film, annealed at 600 °C with second sapphire layer. (c) 200 Å film, annealed at 600 °C with second sapphire layer.
However, the sample shown in figure 5-4 (a) (numbered s317 for future reference) has a significantly larger peak $(\Delta R)/R$ value than any of the other samples. The peak $(\Delta R)/R$ value of $4 \times 10^{-3}$ is four times greater than the $(\Delta R)/R$ values from other samples. The $(\Delta R)/R$ trace can also be converted to a $(\Delta \alpha)/\alpha$ plot to compare with previous results from InAs-doped films deposited directly on sapphire substrates (figure 5-5). It is clear from this plot that this reflective device should have a saturation fluence that is a factor of 2.5 lower than the transmissive device and would therefore be expected to start modelocking.

**FIGURE 5-5:** Comparison of a reflective gold saturable absorber with a transmissive saturable absorber deposited on a sapphire substrate.

This sample was fabricated in the same run as the other samples grown with 400 Å films. During rf sputtering with the non-magnetron system at Lincoln Laboratories, the thickness of the deposited films is dependent upon sample position on the substrate plate. Therefore, differences in the pump probe signals could be due to differences in film thickness between samples that lead to Fabry-Perot etalon effects. This will be discussed in greater detail in a subsequent section.

Based on the experimentally determined absorption saturation dynamics of the reflective gold saturable absorber devices, it is clear that only one of the devices would be expected to self-
start Kerr lens modelocking. The other devices that were fabricated have high saturation fluences
that are not likely to self-start KLM. However, at this point it was not clear why s317 had signifi-
cantly better performance than the other devices, and also why most devices had significantly
poorer nonlinear optical characteristics than previously expected.

5.1.4 Laser Modelocking Experiments

Experiments were done to test the performance of the reflective gold saturable absorber
devices in starting laser modelocking. Samples with both 200 and 400 Å films were placed in the
second fold of the same laser cavity described in chapter 4. The laser cavity was now used in a
reflective geometry, with the 7.5 cm ROC mirror replaced by a reflective gold absorber and the
Brewster angled substrate removed (figure 5-6). Mirror CM2 has a 5 cm ROC and the angle of
incidence is approximately 10°; the angle was as small as possible to introduce a minimal amount
of astigmatism into the cavity. All other cavity specifications were the same as previously
described.

The laser was initially modelocked with a flat high reflecting dielectric mirror in the second
cavity fold. The dielectric mirror was placed at the focus of the second fold, about 2.5 cm from
CM2. Modelocking was optimized to obtain 200 mW output power using a 3.5% output coupler.
The bandwidth was 40 nm at 790 nm center wavelength. When the dielectric mirror was replaced
by reflective gold absorber devices, different laser behavior was observed depending on the partic-
ular device tested. With most samples tested, KLM was obtained with powers in the tens of mW
range, but no self-starting was obtained and no significant increase in laser stability was observed.
However, with the sample that yielded the large pump probe signal (s317, shown in figure 6-4
(a)), a distinct increase in laser stability and robustness was observed; the optical table on which
the laser is mounted could be moved and jarred with no loss of laser modelocking, which is rarely
the case in KLM with no absorber. This indicated that the absorber was affecting laser perfor-
ance, although self-starting was not obtained.
A quantitative measure of this improvement in laser performance can be obtained by measuring the stability region for laser modelocking as a function of the separation between the two folding mirrors CM1 (figure 5-6). Cavity stability regions as functions of both the folding mirror separation and crystal position have been extensively studied [23] [60] [61] and will not be detailed here. The main results of these studies show that an astigmatic laser cavity has two regions where stable lasing can be obtained, measured as a function of the folding mirror separation, and KLM action is maximized on the inner edge of the outer stability region.

In this experiment, the laser was first modelocked with a dielectric mirror in the second fold, as before, with 140 mW output power and 58 nm bandwidth for 5.5 W pump power and 3.5 % output coupling; the stability region was measured by translating one of the folding mirrors in either direction until modelocking was lost. The dielectric high reflector was then replaced by the reflective absorber s317 and stable modelocking was obtained, with 30 mW output power and 44 nm bandwidth. The stability region was measured by the same procedure and found to increase
by a factor of 2. This implies that the absorber was improving laser robustness and stability when compared to a dielectric mirror.

When examining the results of these laser modelocking experiments and correlating them with pump-probe experiments, some trends can be observed. Pump probe signals were obtained with all devices incorporating 400 Å films; however, KLM was not obtained with all these devices. A correlation can be made between the magnitude of the \((\Delta R)/R\) signals and the laser modelocking results for these devices. Modelocking was obtained with samples having \((\Delta R)/R\) signals greater in magnitude than \(-1.5 \times 10^{-3}\), but could not be obtained with other samples. This suggests that these saturable absorber devices were helping start and sustain modelocking, since without sufficient saturable absorber action the linear absorption in the cavity was too large to sustain KLM.

Pump probe experiments with the 200 Å films also offer some interesting insight into laser modelocking performance. Pump probe signals could not be obtained from all of these devices; however, KLM was obtained with all 200 Å films that had detectable signals. These samples provided enough saturable absorber action to overcome the loss introduced by their linear absorption and sustain KLM. The pump probe results indicate that the nonlinear loss modulation introduced by the 200 Å films is actually weaker than that from most 400 Å films, even those that did not start modelocking. However, since the linear absorption was lower in these devices, KLM action would be stronger in the laser cavity and require less saturable absorber action to sustain modelocking. Therefore, the interplay between the saturable absorber action and linear absorption introduced by devices with different film thicknesses seems to govern the modelocking dynamics.

At this point it was still unclear why the majority of the devices did not have the expected decrease in saturation fluence. It was also not understood why s317 had significantly different dynamics than other devices grown under the same conditions. Finally, although s317 clearly improved modelocking performance, it was expected to self-start laser modelocking based on the experimentally determined nonlinear optical characteristics. These issues will be addressed in the next section.
5.1.5 Fabry-Perot Etalon Effects

Fabry-Perot etalon effects were investigated to determine whether they were responsible for the differences in nonlinear dynamics between devices and the problems with modelocking performance. These effects in general occur when a Fabry-Perot etalon is formed by two partially reflecting surfaces separated by a certain distance. The amounts of reflection and transmission depend on the distance between the surfaces and the wavelength of the incident light. The cavity exhibits resonances at certain wavelengths; at these resonances the transmission through the whole structure is a maximum, the reflection is a minimum, and the circulating intensity in the cavity can be many times the incident intensity. These etalon effects are well known and comprehensively described in [16]. They were neglected during the device design in section 5.1.1, when only the standing wave pattern near the gold mirror was considered.

For the reflective absorber devices described in this section, a Fabry-Perot etalon is formed by the two sapphire spacer layers and the InAs-doped silica film, on top of the gold mirror. The first mirror of the etalon is formed by the interface between the first sapphire spacer layer and air (with an intensity reflectivity of 7%) and the second mirror is formed by the 97% reflective gold mirror. These structures are similar to the A-FPSA devices previously discussed in section 3.1.1 and can be modeled in an analogous manner. These calculations were made to determine whether variations in thickness between devices due to differences in position on the substrate platform within the rf sputtering system could have an effect on device performance.

Modeling was done based on the treatment in section 3.3 (a) of reference [62]. The parameters to be calculated were the reflectivity, the intensity at the absorber layer, and the group velocity dispersion, all as functions of both wavelength and thickness of the etalon. In this model, the two sapphire layers and the InAs-doped film are taken to be one layer having the refractive index of sapphire, n=1.7, and the absorption introduced by the film was neglected. The electric field in the etalon is given by

\[ E = A_1 e^{ikz} + B_1 e^{-ikz} \]  \hspace{1cm} (5-3)
The coefficients $A_1$ and $B_1$ can be found by matching boundary conditions at the two interfaces (air-sapphire and sapphire-gold). They are found to be

$$A_1 = \frac{2E_0}{(1 + p_{01})(R_{01} + \frac{1}{R_{12}}e^{-2ikL})} \quad (5-4)$$

$$B_1 = \frac{A_1}{R_{12}}e^{2ikL} \quad (5-5)$$

where $R_{01}$ and $R_{12}$ are the field reflectivities at the interfaces between air and sapphire and sapphire and gold, respectively. $E_0$ is the incident electric field and $p_{01}$ is the ratio of the indices of refraction between sapphire and air. The reflection coefficient can also be found from this analysis and is given by

$$R = \frac{R_{01} + R_{12}e^{2ikL}}{1 + R_{01}R_{12}e^{2ikL}} \quad (5-6)$$

which is equivalent to the expression derived in [62]. Finally, an expression for the group velocity dispersion of a Giles-Tournois interferometer, which is essentially the same as this structure if the gold mirror is assumed to have 100% reflectivity, can be found in [22] and is

$$GVD = \frac{d^2}{d\Omega} \Psi(\Omega) \bigg|_{\omega_i} = \frac{2R_{01}((R_{01})^2 - 1)\sin\delta}{1 + R_{01} - 2R_{01}\cos\delta} \quad (5-7)$$

where $\delta$ is the phase shift due to the propagation between the two mirrors. Equations (5-4) and (5-5) were substituted into (5-3) and the expression was evaluated at the center of the etalon, where the absorber layer is located. This treatment extends the analysis of section 5.1.1 to include all reflections within the etalon.

The intensity was plotted as a function of wavelength for a constant etalon thickness of 254 nm (the design value) and as a function of etalon thickness for the operating wavelength of 800 nm. Equations (5-6) and (5-7) were also plotted in the same manner. The plots as a function of wavelength are shown in figure 5-7, and as a function of thickness in figure 5-8.
Reflection from a Fabry-Perot etalon; \( \lambda = 254 \) nm

Intensity at center of etalon; \( \lambda = 254 \) nm
FIGURE 5-7: Calculations of (a) reflectivity, (b) intensity at the InAs-doped silica film absorber layer, and (c) GVD of the reflective gold absorber devices as a function of wavelength, for an etalon thickness of 254 nm.

Using the results of these simulations as well as the methods described in section 3.1.1 for designing an A-FPSA [35], several observations can be made. Equation (3-1) gives the condition for antiresonance; for resonance, the total phase shift would instead be a multiple of $2\pi$. The design for the reflective gold absorber devices incorporated a total phase shift of $4\pi$ when including the $\pi$ phase shifts at the front and back surfaces, indicating that the etalon is operated at a Fabry-Perot resonance. At resonance, the intensity in the etalon is high and reflection is a minimum. This agrees with the plots shown in figures 5-7 and 5-8 (a) and (b). It should be noted that the maxima and minima in all plots do not occur exactly at the predicted values of 254 nm thickness and 800 nm operating wavelength; the refractive index of the saturable absorber layer is taken to be that of sapphire instead of SiO$_2$, leading to small errors in the plots. In this case, operation at resonance is desired to increase the intensity on the absorber and effectively lower the high saturation fluence of the InAs-doped silica films. For a high-finesse structure, the high
reflectivity of the first mirror makes it very hard to operate at resonance since a small variation in etalon thickness leads to operation at antiresonance. However, for the reflective gold absorber devices the low reflectivity of the first surface relaxes the design tolerances and allows operation at resonance.

(a)

(b)
FIGURE 5-8: Calculations of (a) reflectivity, (b) intensity at the InAs-doped silica film absorber layer, and (c) GVD of the reflective gold absorber devices as a function of thickness, for an operating wavelength of 800 nm.

The increased intensity at the absorber layer would increase the linear absorption of the device; this combined with operation at resonance would reduce the reflectivity of the devices and make operation in the laser cavity more difficult. This could explain the low reflectivity that was measured with the spectrophotometer. It can also be seen from figure 5-8 (b) that variations in thickness would reduce the intensity seen by the absorber layer and thereby decrease the signal measured in pump-probe experiments. This supports the hypothesis that variation in thickness between samples led to difference in measured pump-probe signals and observed modelocking performance; however, the thickness variation required would be on the order of hundreds of angstroms and it seems unlikely that variations due to position on the substrate platform in the sputtering system would be this large. Although not completely satisfactory, this appears to be the best explanation to date for the difference in absorption saturation dynamics between samples.

The GVD of the reflective absorber devices was characterized both theoretically (figures 5-7 and 5-8 (c)) and experimentally using white light interferometry, a technique described in [63]. It
is clear from the calculations that the GVD of these devices is relatively small and should therefore have little effect on laser modelocking. The experimental results agreed with these calculations. At this point, it is still unclear why the sample possessing a large pump probe signal did not self-start Ti:Al$_2$O$_3$ laser modelocking, and further investigations will be performed in the future.

5.2 Reflective Saturable Absorber Devices on a Dielectric Mirror

Another step in the development of reflective saturable absorber devices using semiconductor-doped silica films would be to fabricate reflective saturable absorber devices by depositing films on a dielectric mirror. This would open up the possibility of depositing semiconductor-doped films on dispersion compensating devices such as double chirped mirrors, enabling both a starting mechanism and dispersion compensation in a single device. If successful, this technology could make modelocked lasers significantly more compact and robust, opening up a wide variety of applications.

5.2.1 Fabrication of Dielectric Mirror Absorbers

300 Å thick InAs-doped silica films were initially deposited on standard flat highly reflecting dielectric mirrors. The devices had good optical quality after deposition using the non-magnetron rf sputtering system. However, after a rapid thermal annealing treatment at temperatures from 500-700 °C for 60 s the dielectric mirrors showed a significant degradation in optical quality. The surfaces of the mirrors were essentially unusable, with cracks and pieces of the surface breaking off. To use these dielectric mirrors an alternative method of controlling the absorption saturation dynamics of the semiconductor-doped silica films would have to be developed.

An alternative approach was to find mirrors that could withstand the high annealing temperatures. Specially fabricated high reflecting mirrors obtained from Coherent, Inc. were made to withstand high temperatures and preserve optical quality. To test this experimentally, the mirrors were annealed at temperatures between 400 and 750 °C for 60 seconds. Visual inspection confirmed that the reflection properties of these mirrors should still be suitable for laser modelocking. Linear transmission measurements will be described in the next section.
According to Coherent, the mirrors were designed to have a peak in the electric field located at their surfaces. This implied that InAs-doped films could be directly deposited on InAs-doped silica films, with no spacer layers, and the saturation fluence would be reduced by a factor of 4 as in the design for reflective gold absorbers. 400 Å InAs-doped films were deposited on these mirrors and subsequently annealed at temperatures between 500-750 °C. The mirrors retained optical quality after the deposition and RTA processes.

5.2.2 Linear Optical Properties

The Cary 5E spectrophotometer could not be used to measure the linear reflection properties of the dielectric absorber devices, since the Coherent mirrors were only 1/4” in diameter and would not fit in the reflective attachment. The mirrors could be measured in transmission using the spectrophotometer. The linear transmission spectra before film deposition were found to be essentially unaffected by a rapid thermal annealing treatment, agreeing with visual observations. When 400 Å films were deposited on the surface of the devices, the transmission of the mirrors was unchanged through the center high reflecting band between approximately 700 and 900 nm; however, the characteristic oscillations in the reflectivity for longer wavelengths changed in frequency, indicating a possible shift in layer thicknesses (see figure 5-9). The magnitude of the transmission also decreased for shorter wavelengths due to the increasing absorption of the InAs-doped silica films.

Reflection measurements were performed using a standard Ti:Al₂O₃ laser operating at 800 nm with 260 mW output power. The devices were first glued to a 1” diameter silver mirror in order to mount in a standard mirror holder. This mounting scheme also made it simple to test different absorber devices by rotating the mirror to another device, eliminating the need to remove the device from the mount. When reflection was measured at this wavelength, it was found to be much lower than the expected value of 88% (3% single pass through the film, multiplied by 4 since the intensity is a factor of 4 higher at the surface of the mirror). The power reflected from the mirrors ranged between 62 and 73%. This reduction in reflectivity was independent of the RTA treatment, since both samples that were annealed and samples that were not annealed had similarly low reflectivities. These absorptions would probably be too large to permit lasing in the Ti:Al₂O₃ cavity.
Several explanations were suggested for the observed decrease in reflectivity. One possibility was that the deposition rate of the InAs+SiO₂ target in the rf sputtering system had increased and therefore the deposited films were thicker than expected. This hypothesis was tested by recalibrating the deposition rate for this target; it was found to be 20 Å/min as before. Another possibility was that the dielectric mirror surfaces were affected by the exposure of the substrate to the plasma for substrate cleaning before the films were deposited. A sample was fabricated without substrate cleaning and with a thinner 200 Å film to introduce less absorption into the laser cavity. This sample reflected approximately 80% of the incident energy at 800 nm, which was still significantly lower than expected; however, this could be a small enough absorption value to enable lasing. The reflectivity was found to vary from 70-83% over the wavelength range from 790 to 850 nm, with approximately the same shape as the transmission curve for absorbers grown directly on sapphire substrates.
5.2.3 Pump Probe Experiments

The nonlinear optical properties of the reflective dielectric saturable absorber devices were measured using the pump probe system in the reflective configuration described in section 3.1.3. Some representative traces are shown in figure 5-10.

![Graphs showing pump probe traces](image)

**FIGURE 5-10**: Pump-probe traces from InAs-doped silica films deposited on dielectric mirrors. (a) 400 Å InAs-doped films annealed at different temperatures. (b) Comparison of 200 Å and 400 Å InAs-doped films, both annealed at 600 °C; baselines are subtracted out.
Some trends can be observed from these traces. When examining the 400 Å films as a function of annealing temperature (figure 5-10 (a)), it appears that the magnitude of the \((\Delta R)/R\) signals increased with increasing annealing temperature, implying a decrease in saturation fluence. In particular, devices annealed at 750 °C exhibited a signal that is significantly higher than that of InAs-doped films deposited in a transmissive configuration, as expected from the design. This trend is somewhat different than that seen for InAs-doped films deposited directly on sapphire substrates, for which the signal magnitude remained approximately constant as annealing temperature was increased. The decrease in the baseline for these devices does agree with the dynamics observed with transmissive devices. The relaxation dynamics also were faster for devices annealed at higher temperatures, which agrees with trends seen in the previous data on InAs-doped films [13]. These trends are not conclusive, however, since experiments were only performed on a small group of mirrors. These samples appear to have a large enough nonlinear loss modulation to start laser modelocking; however, as previously discussed, the loss introduced into the cavity may be too large to sustain lasing.

The dynamics of the sample fabricated with a 200 Å film and annealed at 600 °C can also be compared to the dynamics of the other reflective absorbers with thicker films, particularly the absorber with a 400 Å film annealed at 600 °C (figure 5-10 (b)). The absorption saturation dynamics are very similar for both devices when comparing both the signal magnitude and the relaxation dynamics. These observations seem to indicate that the RTA treatment affects devices in the same manner regardless of film thickness, as expected from the earlier work on transmissive devices. However, although this device may introduce small enough linear absorption to sustain lasing, the magnitude of the pump probe signal is probably too small to self-start laser modelocking.

5.2.4 Laser Modelocking Experiments

The reflective dielectric absorber devices were applied to modelocking of a Ti:Al₂O₃ laser. The cavity configuration was identical to the layout previously described in section 5.1.4. The laser was modelocked with a standard dielectric mirror in the second fold as before. The dielectric mirror was replaced first with a reflective dielectric absorber device incorporating a 400 Å InAs-
doped silica film. However, with this device in the cavity it was impossible to obtain lasing. Several devices with 400 Å films and annealed at different temperatures were tested in the laser cavity; however, all of them introduced too much loss and prevented lasing, as expected from the previous linear absorption measurements. The device incorporating a 200 Å film was then introduced into the cavity and lasing was obtained.

With this device, KLM was obtained with an output power of 150 mW with a 3.5% output coupler and 5.5 W pump power. The pulses had 37 nm bandwidth at 810 nm center wavelength. KLM was easier to start with this device, but self-starting was not observed. The output coupling and pump power were systematically varied in an attempt to obtain self-starting modelocked operation. However, although it appeared the device had some effect on laser modelocking, self-starting modelocking could not be obtained. This agrees with the pump probe measurements, since this sample had a \((\Delta R)/R\) signal of only \(1.5 \times 10^{-3}\).

The modelocking results and pump probe results for this reflective dielectric absorber were quite similar to those of the reflective gold absorber with pump probe characteristic shown in figure 6-4 (b). The modelocked output powers and bandwidths obtained from both devices also correspond well; 90 mW and 42 nm bandwidth was obtained from the reflective gold absorber and 100 mW with 37 nm bandwidth from the reflective dielectric absorber for 2% output coupling and 5.5 W pump power. This further substantiates the explanation of the modelocking dynamics advanced in section 5.1.4; the interplay between saturable absorption, measured in pump probe experiments, and loss introduced into the cavity, proportional to output power, should govern the laser modelocking dynamics. From this qualitative explanation, the devices would be predicted to behave similarly when introduced into the laser cavity, which is borne out by experimental results in this case. This indicates that knowing both the pump probe characteristic and the linear absorption of a given device should allow a qualitative prediction of the modelocking dynamics in the laser.

From these experiments, it is clear that further work needs to be performed to make these reflective dielectric mirror absorbers self-start modelocking. The remaining difficulties include explaining the reduced reflectivity of the devices after films are deposited and also explaining the
difference in the effect of annealing between these devices and transmissive devices. Since the devices annealed at 750 °C had a significantly larger pump-probe signal than other devices, it seems possible that devices with thinner InAs-doped films and annealed at 750 °C would have a small enough absorption to sustain lasing and a large enough nonlinear loss modulation to self-start modelocking. Experiments aimed at clarifying these effects will be carried out in the near future.
Chapter 6

Summary and Conclusion

This thesis has discussed the further development of nonepitaxially grown saturable absorber devices. These devices, consisting of semiconductor nanocrystallites doped into silica films grown using rf sputtering and then undergoing rapid thermal annealing, offered several advantages over epitaxially grown absorbers. These included inexpensiveness, almost unlimited choice of absorber and substrate materials, and ability to control the absorption edge and linear absorption introduced by the device. Previously, these devices were shown to be very successful in modelocking Ti:Al₂O₃ lasers, resulting in self-starting 25 fs pulses that were tunable from 800 to 880 nm. The primary difficulty with these devices was their high modulation depth, making it impossible to modelock the laser without the aid of KLM.

Three distinct approaches were taken in advancing this novel technology. The first approach was to develop magnetron rf sputtered GaSb-doped silica films. This was expected to lower the high saturation fluence of the saturable absorber films. With magnetron sputtering, films were expected to have higher quality and also the nanocrystallite sizes were expected to be larger, resulting in a lower saturation fluence. In linear transmission experiments the absorption edge was found not to vary with rf power or film thickness. Initial pump probe experiments done on samples before annealing indicated that the saturation fluence of these devices was lower than in the previous work. However, the rapid thermal annealing process significantly reduced the optical quality of most GaSb-doped silica films, making it impossible to obtain a pump probe signal from almost all samples tested; samples that did show a pump probe signal showed no decrease in saturation fluence. Laser modelocking experiments confirmed these predictions, as self-starting KLM was not observed and modelocking performance was generally unaffected by the introduction of the GaSb-doped films in a transmissive geometry.
With GaSb-doped films grown in the magnetron sputtering system at a substrate temperature of 100 ºC and subsequently annealed, oscillations in the pump probe signal were observed. These oscillations were attributed to coherent phonons. A simple theoretical model was applied to show that these were not due to the lowest order acoustic phonon; further investigations are needed to explain this phenomenon.

The second approach was to incorporate the InAs-doped silica films deposited using the non-magnetron sputtering system from the previous work in a reflective geometry. By placing the saturable absorber film between two transparent spacer layers, the effective saturation fluence of the device could be controlled. Devices were successfully fabricated using sapphire spacer layers after initial problems with annealing devices that had silica spacer layers. Linear reflection measurements showed lower reflectivity than expected. Nonlinear optical measurements revealed that only one of the devices would be expected to start laser modelocking, since it had the expected factor of four decrease in the saturation fluence. However, when incorporated into a laser cavity, none of the devices self-started modelocking.

Fabry-Perot etalon effects were analyzed theoretically to determine whether they were responsible for the differences in absorption saturation dynamics between samples and also the inability of the device that had a lower saturation fluence to start modelocking. It was found that the devices, if fabricated with the correct thickness, should operate at a Fabry-Perot resonance and therefore have reduced reflectivity but the expected factor of four intensity enhancement at the absorber layer. If the thickness varied significantly between devices, the intensity on the film would drop and saturable absorber performance would be negatively affected. The group velocity dispersion of the devices was also analyzed and found not to affect modelocking performance.

Therefore, at this time it appears that variations in etalon thickness between devices are responsible for difference in absorption saturation dynamics. To determine this conclusively, experiments could be done to characterize difference in film thickness across the substrate platform in the non-magnetron sputtering system using photolithography. It is also still unclear why the sample with a lower saturation fluence did not self-start laser modelocking, and further investigations must be done to determine this.
Finally, InAs-doped silica films were deposited on dielectric mirrors using the non-magnetron sputtering system. Initially, standard dielectric mirrors did not survive the annealing process. However, films were deposited on specially designed dielectric mirrors from Coherent and the whole structure was successfully annealed. The reflectivity of these devices was lower than expected, for reasons that are as yet unclear. Pump probe experiments on these devices revealed that samples that had a large enough signal to start modelocking had too low a reflectivity for use in the laser, and samples with high enough reflectivity had saturation fluences that were too high to self-start modelocking. This trend was borne out by laser modelocking experiments, where the samples had some effect on modelocking but self-starting was not obtained. In general, for the reflective absorber devices on both gold and dielectric mirrors, tradeoffs between saturable absorber and KLM action were observed in modelocking dynamics.

Many future directions can be taken with this work. The magnetron sputtered GaSb-doped silica films looked promising before the RTA treatment; therefore a solution would be to use the magnetron sputtering system with InAs+SiO₂ targets, since these films have previously been shown to withstand the annealing process. Control of the substrate temperature and improved deposition rates and film quality should reduce the saturation fluence when compared to the films grown without a magnetron system, although the coherent phonon effects observed with GaSb-doped films after annealing may affect these samples in a similar manner when grown with a certain substrate temperature.

Another possibility is to use InAs-doped silica films deposited on dielectric mirrors and annealed at 750 °C, which exhibited a low saturation fluence in pump probe measurements. This did not work before because the 400 Å films introduced too much absorption; however, using 200 Å films could overcome this problem. Other directions taken from this work in particular include determining why the reflective gold absorber devices with a lower saturation fluence did not modelock the laser, and also to fabricate and apply reflective absorber devices with magnetron sputtered films.

General areas of future work include extending this technique to other solid state laser systems by either controlling the quantum dot size or changing the semiconductor dopant in the sputtering system. Another way to control the saturation fluence as an alternative to annealing could
also be found in order to surmount problems with semiconductors that lose optical quality after an RTA treatment. This would also enable film deposition on standard dielectric mirrors. Overall, this is a very promising technology that has the potential to make saturable absorber devices much more flexible and easily fabricated, leading to more compact, robust, and inexpensive ultrafast laser sources.
References


