Statement

I hereby declare that this submission is my own work and to the best of my knowledge it contains no materials previously published or written by another person, or substantial proportions of material which have been accepted for the award of any other degree or diploma at UNSW or any other educational institution, except where due acknowledgement is made in the thesis. Any contribution made to the research by others, with whom I have worked at UNSW or elsewhere, is explicitly acknowledged in the thesis. I also declare that the intellectual content of this thesis is the product of my own work, except to the extent that assistance from others in the project's design and conception or in style, presentation and linguistic expression is acknowledged.

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ABSTRACT

Porous silicon (PSi) has been shown to be a material with varieties of nonlinear optical properties. These properties have been discussed in terms of its nature of photoluminescence mechanism such as quantum confinement, surface states and the role of impurities. PSi has also been shown to be a useful material for optical devices, and have potential applications in fields such as bio-sensing and efficient lighting. In the following thesis, I present my theoretical and experimental results in studying a nonlinear optical property, known as optical bistability, in mesoporous silicon microcavity devices.

The results show significant optical hysteresis in the transmission and reflection properties of mesoporous silicon microcavities when illuminated with a 150 nanosecond pulsed laser at 532 nm. The optical hysteresis is shown to be transient in nature and the properties are strongly dependent on the porosity of the cavity layer. The onset and damage threshold intensity are also shown to be porosity dependent. Modeling suggests that the observed effects are due to changes in the nonlinear refractive index and the transient lifetime increases with increasing porosity. The role of surface states was also investigated on influencing the bistable process by passivating the internal porous surface with hydrosilylation chemistry.

The role of the number of periods in the cavity structure is also studied. The cavity is doped with quantum dot to improve its switching intensity. The optical bistable property of the cavity was also investigated by using a 532 nm continuous wave (CW) laser with maximum power of 10 mW, modulated by an optical chopper. The only sample that yields significant hysteresis is the sample doped with quantum dot, with millisecond response time. The results
show that the sample doped with quantum dot shows both electronic and thermal process in forming optical hysteresis.

Finally, I develop a theoretical model to study the transient hysteresis shape. The model suggests that the power and pulse width of the excitation laser source strongly influence the shaping of the bistable curves. The nature of the nonlinear refractive index is also quantified in terms of the power and the width of the excitation pulse. The third order nonlinear coefficient is calculated to be in the order of $10^{-10}$ esu while the passivated samples yield results in the order of $10^{-9}$ esu, which agree with previous findings in literature.
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This project has been carried out in the Optoelectronics Laboratory in the School of Physics at the University of New South Wales, Sydney. This work would not have been possible without the generous assistance of many other people whom I have been working with during my study, and it is these people which I would like to express my gratitude.

First and foremost I would like to thank my supervisor Dr. Peter J Reece who has provided me continual guidance and support during the course of my project. His vast scientific knowledge and enthusiasm have helped me so much in understanding my research and overcoming many difficulties while working on this project, as well as in training me to be a scientist in the future. I am very grateful for the opportunity to have worked and learned from him. Secondly, I would like to thank my co-supervisor Prof. Michael Gal for his insightful suggestions and wise advices during my research and my future study. I wish him all the best for his retirement.

I also would like to express my sincere thanks to Hong Qiao who helped me with my understanding of porous silicon and providing me with samples for my research. I am truly indebted to Hong’s patience in keeping up with my demands of “fresh” samples, and without his help, this project would not have been completed. I must also thank Bin Guan for her help in doing surface passivation for my samples. Her vast knowledge in the topic has benefited me a great deal in understanding the process.
To all the members of the Optoelectronics group I express my heartfelt thanks to. I have been lucky enough to work in a warm and welcoming environment. To Fan Wang, my friend and colleague, it has been an honor to share the same office with you during the last few years. Even though he is always busy with his research, we spent quite a lot of time together discussing many aspects of optics. I will never forget many conversations we had in the office and during the lunch times, they have been very entertaining and useful. I wish him all the best with his PhD. To Wen-Jun, my partner in crime in playing “Quake”, thank you for entertaining me with your game skill, and all the best to your future. To Craig, who has provided stimulated discussions in many different topics in society, it has always been very interesting to share my ideas with you, and good luck with your research. To Kelly, Tim and Anna, I wish you all the best with your study. Thanks also to Patrick McMillan who taught me that the best way to understand science is to be able to explain it to anyone in plain terms.

On a personal note, I would like to thank my friends and “comrades in-arms”, Lian, Robert and Jolyon. I am indebted to them above all else. Without many hours of stimulating conversations with them, I do not think I would have been sane enough to complete this research project. They have always been there for me in time of needs. Finally, I would like express my deepest gratitude to my parents for all their financial and emotional support during my course of study.
I. INTRODUCTION

Porous silicon (PSi) has been studied extensively for its optoelectronics properties since the observation of strong visible photoluminescence in 1990 [1]. Initial research focused on light emitting devices [2], however a wide range of integrated optical components have also been investigated, including photodetectors [3], waveguides [4], and modulators [5]. In many of these early devices, optoelectronic properties were linked to the presence of a complex and extensive nano-structured internal surface which permeates throughout the material. The identification of the exact physical mechanism of observed effects, such as strong visible and near-infrared luminescence, was complicated by conflicting experimental results that have been alternately ascribed to the quantum size effects [6-8], surface defects [9-10] and the presence of impurities [11]. Whilst this still remains a topic of debate [12], it is clear that the surface plays an important role in mediating many of the observed effects [13].

The ability to create multi-layered photonic structures from PSi, first demonstrated by Berger et al. [14], provides a further dimension to device design with a host of passive optical components and active devices possible [15-16]. By controlling the degree of porosity of the PSi films during the etching process, the resulting refractive index of the layer may be tuned from approximately 1.3 to 2.6. As the electrochemical reaction is a self limiting process, regions of different porosity may be etched beneath existing PSi layers to create one-dimensional planar photonic structures. Demonstrated passive optics components include high-finesse microcavity resonators [17], broadband dielectric laser mirrors [18], graded index lenses [19] and Rugate filters [20]. Notably, the presence of the interconnected porous network also provides an alternative means of achieving functionality in PSi devices, by incorporating foreign materials into the porous matrix to create hybrid devices. In terms of active devices, resonant modes have
been used to enhance intrinsic light emission from PSi and other optically active dopants (e.g. rare earth ions [21], Rhodamine dyes [22], colloidal quantum dots [23]), whilst periodic Bragg reflectors have been used to enhance nonlinear optical effects of second (SHG) and third harmonic generation (THG) [24-26].

Another class of optical devices which benefits from the use photonic structures is in the area of optical bistability. Optical bistability refers to systems that exhibit two stable output states for the same input light intensity when the input is varied over a range of values, and as such have potential applications in optical computing and information processing, i.e. optical memories and fast switching [27]. Optical bistability is usually studied by characterizing the transmitted or reflected intensity as a function of input intensity in hysteresis-like curves by rapidly switching of the excitation source.

Recent studies have demonstrated that PSi can also exhibit strong optical bistability. Initial work focused on single 16-25 μm thick films of mesoporous silicon at 593 nm [28], whilst the first PSi microcavity bistable devices operating at a wavelength of 800 nm were demonstrated by Takahashi et al. [29]. In these studies the bistability was shown to be transient in nature and the mechanism of switching has been thought to be linked to phonon confinement [28]. More active structure such as porous silicon photonic filled with condensate gas has been able to show much sharper switching [30] with very low optical power input.

In most optical bistable curves, the hysteresis shows sharp switching between the input and output intensities which presents a potential for a potential application in terms of making a new form of bio-sensor without the need of a spectrometer. PSi multilayered structures have demonstrated their abilities of sensing in a variety of ways from one dimensional photonic
structures such as Bragg reflectors [31] and Rugate filters [32] to microcavity structures [33]. However all of these sensors require monitoring the shift in the resonance peaks using a spectrometer. Consequently, a study in optical bistability in mesoporous silicon microcavities is a possible novel form of sensing by modulating the input light and read out the results from an oscilloscope.

In the following thesis, I will present key findings in the study of optical bistability using mesoporous silicon microcavity devices. The contents of thesis are organized in the following way:

i) **Theoretical background** describes an overview of optical bistability such as the long history of the subject as well as various forms of materials which have been studied. Various applications of optical bistability is also mentioned, and a basic treatment of the common physical mechanisms in optical bistability along with the mathematical modeling of the steady state process. A summary of research done on porous silicon as a potential material for optical bistability is described in details. The second part of the theoretical background deals with the actual mathematical model used to describe the transient process recorded in the experiments. This involves a detailed analysis of a Fabry-Perrot cavity with linear absorption loss and filled with nonlinear refractive index material.

ii) **The Experimental method** concerns the fabrication of the porous silicon structures. A summary of the anodization process is mentioned as well of the surface passivation process using hydrosilylation chemistry. The optical bistability experimental setup and the transmission/reflection study are described in details with explanation of different instruments used in the course of the study.
Two methods used to study optical bistability at 532nm are discussed. One is used to understand the fast response by using a nanosecond pulse, and the other one is for studying the thermal process by using a continuous-wave diode laser.

iii) *Results* section discusses findings during the experimental process. Four key topics are presented. First, a summary of the transmission/reflection results is presented with analysis of the cavity structures such as the linewidths and the FWHM. Second, it shows the optical hysteresis process in empty cavities with (A) a range of porosities as the central layer and (B) with different surface passivation conditions. Third, the section describes the fast and slow (thermal) process happening in sample doped with 550 nm quantum dot and lastly the influence of the number of mirrors layers on the bistability performance of the cavity structure.

iv) Detailed analysis of the bistability process in the empty cavity structures are studied in *Discussions*. The results of the fitting process such as the transient lifetime and the nonlinear coefficient are presented. A detailed study of the influence of the excited pulsewidth and the nonlinear refractive index are also discussed in detail.

v) Finally, a *Conclusion* of all the experimental findings is summarized and several suggestions are made to improve the understanding of the bistability process in the devices.
II. THEORETICAL BACKGROUND

1. Optical bistability

2.1.1 Introduction to Optical Bistable Devices

Optical bistable devices are system that exhibits two stable outputs with the same input intensity over a range of input values. The most common optical bistable device is the Fabry-Perot (FP) cavity with an intensity-dependent nonlinear gain medium. By placing the nonlinear material inside the FP cavity, reflectivities from the mirrors can create optical feedback, which can amplify the intensities inside the cavity to provide differential gain and hysteresis. Such a feedback system is called intrinsic because it is an all-optical device, and if the feedback is channeled through an electronic circuit, it is called hybrid.

Optical bistability is usually studied by characterizing the transmitted or reflected intensity as a function of input intensity in hysteresis-like curves by rapidly switching of the excitation source. Study of optical bistability in FP cavity filled with nonlinear medium was first conducted by Szoke et al. The medium was SF$_6$ gas and the device was excited by a CO$_2$ laser at 10.6 μm [34]. However, the device only showed nonlinear behavior but did not sharp switching bistable properties. Other attempts were made by Austin and De Shazer by studying the transmission of a ruby single-mode pulsed laser passing through a FP cavity containing cryptocyanine in methanol [35]. They showed the existence of pulse narrowing in the presence of the material which gave rise to optical hysteresis. Spiller utilized organic dye solution as a form of nonlinear gain medium inside a FP cavity with length varied from 25-50 μm [36]. As in previous studies, he only observed the existence of nonlinearities and the sample did not show bistability. A few years later, McCall made the first serious theoretical study of FP cavity filled
with nonlinear medium [37], which laid the foundation for Gibbs et al. to develop the first optical bistable FP cavity device. The medium used in this case was sodium vapor [38].

Following the discovery of optical bistability in sodium, Kerr liquids such as CS$_2$ and nitrobenzene were used in the next successful intrinsic optical bistability devices [39]. The constructed device was a FP cavity with 98% reflective plane mirrors separated by 1 cm distance which corresponds to a cavity round-trip time of about 0.1 ns. The cavity was excited with Q-switched single-mode ruby laser. A similar study was also done with liquid crystal material n-p methoxybenzylidene-p-butylanile (MBBA) which shows strong bistability [40]. Other early works in the 70 s saw the demonstration of optical bistable property in ruby, which was also the first solid material to have such a property. Vekatesan and McCall reported optical bistability using a FP cavity containing ruby between 85 and 296 K [41-42].

Semiconductors were shown to be a viable source of bistable material with the fabrication of a Fabry–Perot etalon that consisted of thin-film GaAs epitaxially grown [43]. Material with similar property like GaSe [44] was also reported to have bistable property at low temperature of 80 K with 50 ns switching times at 592.5 nm excited laser source whose power was 200 kW/cm$^2$. Other subsequent studies in bulk single crystal structures like InSb [45] and InAs [46] also exhibit optical bistability at low temperature. Material like CuCl can demonstrate optically bistable property at 14 K [47] with sub-picosecond switching time using the biexciton two-photon resonance [48]. Intrinsic optical bistability has also been observed in semiconductor ferroelectronic material like SbSI using a 2 mW He-Ne laser source [49].

Studies in optical bistability at room temperature became more of a focus with the observation in HgCdTe [50] due to induced two photon absorption. Kar et al. demonstrated that resonator with 250-μm-thick etalon InSb can also cause optical bistable hysteresis at room
temperature by the same mechanism when illuminated with CO$_2$ laser operating at 9.6 – 10.6 μm [51]. Bistability can also be observed in GaAs at room temperature with low exciting power and wider hysteresis loop [52]. GaSe device is also tested at room temperature with the hope of exhibiting bistability due to its similar property with GaAs, but the hysteresis only shows switch-on and no sign of switching off [53]. Epitaxially grown quantum heterostructures such as quantum wells have also demonstrated the ability to exhibit optical bistability at room temperature with low excited intensity [52]. The large nonlinearities of multiple quantum well structures at room temperature allow the use of diode laser as a light source for bistability [54].

The discovery of optical bistability in semiconductors led to the construction of novel forms of cavities and also of bistable devices without resonators. The advances in the fabrication process have led to the construction of bistable devices in micro-ring resonators [55-56] as well as in photonic crystals [57]. The first resonatorless optical bistable device was predicted by Kaplan who suggested that bistability can be formed by the reflection of light near the critical angle from an interface separating a linear and a nonlinear medium [58-59]. Two years later, Smith et al. [60] reported on the first optical device operated based on the intensity-dependent total internal reflection. Russian researchers Karpushko and Sinitsyn reported on the existence of optical hysteresis in interference filter containing ZnS as the intermediate layer whose thickness is 0.22 μm [61]. Research done on commercial interference filter has been able to show optical bistability with only millisecond switching times but required power as low as 6 kW/cm$^2$ at 514.5 nm [62]. Results in fabricated filters with ZnS or ZnSe as the high-refractive-index layer show faster operation in the order of less than 10 μs switching times, and milliwatt onset power [63]. Other form of resonatorless bistability was proposed by Bjorkholm based on self-focusing [64]. The device operates based on the principle that a laser beam is focused onto the input face
of a medium having an intensity-dependent-index of refraction. The spot-size of the beam is altered when the input intensity reaches certain value. The output power is imaged by the lens onto an aperture which blocks or transmits the beam based on beam spot size after passing through the nonlinear medium. Another type of device operates based on the effect of intense short laser beam propagating in a heterogeneous intensity-dependent medium [65]. This is because when high-intensity pulse propagates through the medium, changes in the pulse inside the material can be caused by light scattering, which is dependent on the topology of the material. Another scattering mechanism is due to the varying dependence of the index of refraction of the heterogeneous medium components on the light intensity for unchanging topology [66].

Other interesting types of intrinsic optical bistable device have also been suggested with the observation of bistability in other materials. One example is the usage of the Doppler-free two photon dispersion near the \(5S_{1/2} - 5D_{5/2}\) in Rb to study optical bistability [67-68]. This was followed by the discussion of an optical tristable system, i.e. a system with three stable outputs from one input source, using two photon mechanism by Herman [69], Walls et al. [70], Tsukada et al. [71], and Agarwal [72]. Cecchi et al. [73] reported on the first observation of optical tristability in Na vapor.

Other types of optical nonlinearities such as degenerate four wave mixing [74-75] and phase conjugation [76-77] have been also proposed to be a good mechanism for optical bistable devices. These types of devices can operate without mirrors, and the feedback is entirely due to the atomic process of the materials. Early works on mirrorless bistability were studied by Bowden and Sung, in which they proposed that the mechanism for the feedback is from the interaction of virtual photon which is called atomic-pair correlation [78-79]. The search for
materials possessing bistability without mirror feedback has led to the discovery of new materials. Materials with helical structure such as chloesteric liquid crystals were predicted to exhibit optical bistability by Greubel [80] and Winful [81], who independently show that the bistable mechanism can arise from the light-induced distortions of the cholesteric helix for travelling light waves. This was later confirmed experimentally by Pura et al. [82] Non-crystalline semiconductors like amorphous Si (a-Si) and chalcogenide glasses (ChG) also show hysteresis like dependent output intensity when excited by strong pulsed laser light [83].

Besides of the use of an all optical system, bistability can also be also achieved through form of hybrid devices where the nonlinearities are synthesized using electro-optic feedback. The first proposal of such a device was by Kastal'skii [84], in which his device was composed of a laser, a Fabry-Perot cavity, a detector, and electrical feedback to an intracavity electro-optic crystal. A similar approach was independently undertaken by Smith [85] and his colleagues [86-87] to demonstrate the bistability of an FP cavity with an electro-optical crystal. The advantages of these devices are that the nonlinearities of the crystal can be driven both optically and electrically so it only requires low power laser. The most successful of these self-electrooptic effect devices has been created by Miller et al [88]. The device combines a modulator operating based on the quantum confinement Stark effect and optical detection within the same structure, creating an optoelectronic feedback that, when positive, gives optical bistability.

These advantages have also led to other efforts of simplifying the bistable devices. One attempt is to create a resonatorless hybrid bistable device, because elimination of the resonator means that coherent light source is no longer needed to create bistability, and if the nonlinearities can be also driven electrically then any forms of light source like LED or incandescent bulb can now be used to provide the excited power. The first proposal was by Marburger et al. [89] and
Garmire et al. [90], Garmires’ device consists of a lithium niobate (LiNbO$_3$) crystal as the source of bistability. Another form of hybrid bistable device is to employ the usage of wave-guide modulators, which has been able to demonstrate by Garmire et al. [91] and Smith et al. [92-93]. The devices consist of a Ti in-diffused waveguide placed on a LiNbO$_3$ substrate. The waveguide is in between 2 parallel electrodes. The ends of the LiNbO$_3$ substrate are cleaved to produce parallel surfaces. Hybrid bistability has also been seen with integrated optical modulator with an induced dielectric channel [94].

Optical bistability not only exists in passive systems, but it also exists in active systems like lasers. The first proposal for the bistable laser was made by Lasher [95] whose device is a FP injection laser. Its p contact is divided into two electrically isolated portions by a slit parallel to the light-reflecting sides of the crystal. However this device is not an actual “optical” bistable system since the input variable is the current level. The actual optical bistability in a laser was not discovered until 1979 [96]. The device studied is a hybrid system consisting of a dye laser with an Ar laser pump source. The laser cavity is tuned by an electro-optical birefringent tuner. The hysteresis is observed as a function input light intensity.

### 2.1.2 Types of Optical Bistability

The optical bistable device proposed by Szoke et al. consisted of a saturable absorber placed inside the Fabry-Perot cavity. They predicted that bistability would occur at resonance if the absorption coefficient of the nonlinear material in the cavity decreased as the input light intensity increases. This property is known as band-filling and was first predicted in semiconductors [97]. When the material is excited by high intensity light, the electrons excited by the input photons can fill the states in the conduction band. This would block further absorption and would result in the material becoming transparent.
Figure 2.1 shows a schematic for a porous silicon microcavity which is composed of two Bragg mirrors and an active layer in between them. Since the optical field passes through the same materials as it enters and exists the active layer, the principle of operation of this device is similar to an optical bistable device based on a FP cavity with a nonlinear medium inside the cavity as described by Smith et al. [98] The output power of the cavity is described by an Airy function which peaks periodically at resonant modes. The material inside the cavity can be a saturable absorber or an intensity-dependant-refractive-index material.

Figure 2.1. (Left) Diagram of a porous silicon microcavity composed of periodic high-refractive-index (dark) and low-refractive-index (bright) layers, and (Right) a FP cavity with two Bragg mirrors M1 and M2, and the active layer acts as a nonlinear medium. An input intensity $I_I$ enters the cavity and exists as transmitted intensity $I_T$ and reflected intensity $I_R$.

For a saturable absorber, if the intensity of light is small, the material absorbs most of the light intensity, which prevents the formation of constructive interference inside the cavity. Therefore the intracavity intensity $I_C$ can be approximated to be proportional to the input intensity $I_I$ as following

$$I_C = TI_I \quad (2.1)$$

where $T$ is the transmission of both the front and back mirrors, and the transmitted intensity $I_T$ is

$$I_T = TI_C = T^2I_I \quad (2.2)$$
The above equation holds when the intracavity intensity $I_C$ is smaller than the saturation intensity $I_S$ [27], i.e., $I_S > TI_I$. When the input intensity reaches certain critical value, the transmitted intensity is comparable to the input intensity, i.e. $I_t \approx I_T$ and $I_C = \frac{I_T}{T}$, due to the existence of saturable absorption inside the cavity. This only exists if $I_C$ is larger than the saturation value $I_S$, so $I_t > TI_S$.

Bistability based on saturable absorption is difficult to achieve [99], and a more common mechanism of optical bistability is based on the nonlinear changes in the refractive index, which is known as dispersive bistability. In a Fabry-Perot cavity, the transmission is dependent on the roundtrip phase-shift, which is also proportional to the refractive index of the material inside the cavity. As a result, if the refractive index has a nonlinear response the cavity can be tuned away from resonance to block light from going through. This mechanism was predicted in 1979 to explain the optical bistability in sodium vapor [100].

Curve A in Figure 2.2 represents the transmission of a FP cavity with intensity dependent refractive index medium at different intensities $I_I$. The curve B represents transmission at certain input intensity. The intersections between the curve A and B correspond to the different switching intensities at certain input intensity. The plot shows that there are one, three, five or even more possible intersections between the two curves corresponding to a system of no bistability, bistability or multiple bistability. For the case when there are three intersections between curves A and B, this plot can be translated into the well-known hysteresis shape as shown in Figure 2.3.

Figure 2.3 represents the input-output hysteresis of bistable system which can be understood in the following way. As the input intensity increases from zero, the output intensity also increases linearly. This corresponds to all the lines whose slope is larger or equal to the line...
B1 in Figure 2.2. These lines only intersect the curve A at one point which means that there is one output for every input, i.e. the relationship between the input and output intensity is linear. As the input becomes larger and reaches certain value $I_c$ as in Figure 2.2, the output suddenly becomes much larger and switches to the upper branch of the hysteresis. If the input keeps increasing, $I_T$ will increase and trace out the path from C-D. If $I_T$ now starts to decreases slowly, $I_T$ will continue to be on the upper branch, until the input reaches certain value of $I_a$, then the output $I_T$ will be dramatically decreases, and the system switches back to the lower branch. Thus, between $I_a$ and $I_c$, two stable states coexist in the same system. By modifying the system parameters, we can obtain devices which have a differential gain or a switching characteristic.

**Figure 2.2.** Transmission of a FP cavity filled with intensity dependent refractive index material. Curve A shows the transmission in the form of an Airy function with intensity dependent optical path length. Curve B1 to B4 show the transmission at different input intensities. The intersections of A and B are the switching points of the hysteresis input-output curve (Adapted from [101])

**Figure 2.3.** Optical hysteresis relationship between the input and output intensity
However, there are certain bistable systems that exert hysteresis but the input-output curves show irregular shapes. These types of bistable curves are usually typical response of a transient system. These transient relationships do not show sharp switch-up or switch-down associated with the hysteresis loop, and there is also no partial overlap between the intensity-increasing pathway (C-D path in Figure 2.3), and the intensity decreasing pathway (A-F path in Figure 2.3). The transient hysteresis has been found to exist in materials such as liquid crystal MBBA in the isotropic phase when excited with 693.4 nm ruby laser [102], and absorptive dye (BDN) solution at 532 nm, and also in non-crystalline materials such as chalcogenide glass (ChG) [103].

Most of these transient curves are usually modeled based on some sorts of trial functions of the nonlinear phase change, and they are always time dependent. Thus, the transient condition is intrinsically linked to the cavity response time $\tau_c$, the material lifetime $\tau_m$, and the time duration of the excited pulse $\tau_p$. There are three different types of transient states which were studied in detail by Bischofberger and Shen [40] to explain the strange hysteresis observed in Kerr liquid medium [39, 102, 104]. The three transient regimes are extremely transient ($\tau_m \gg \tau_p \gg \tau_c$), moderately transient ($\tau_p \gg \tau_m \gg \tau_c$) and quasi-steady-state ($\tau_p \gg \tau_c \gg \tau_m$). It has been suggested that if the thermally-induced refractive index change dominates over other contributions, the observed optical bistable behavior can exhibit a transient nature because of the relatively slow rise time and extremely long relaxation time of the thermal process [105]. Some materials can also exhibit transient hysteresis at different time scales such as in ChG material like As$_2$S$_3$. True and McCaughan [106] have reported light induced refractive index change in thin As$_2$S$_3$ films in low absorption spectral region at both long and shorter time scales. They
demonstrate that for long time scales the thermal refractive index changes are thermal in origin, but for shorter time scales the thermal effect can no longer be observed.

2.1.3 Application of Optical Bistability

An optical system where light is controlled by light has been the long-standing goal for developing all-optical communication components. With the advance of technology, optical ultra short pulses ranging from the order of picoseconds [107], femtoseconds [108], to even attoseconds [109] have been reported. The missing link in an all-optical system is an ultrafast optical switch. Early research has shown that the optical bistable devices have many desirable characteristics in this regard. They can perform a number of logic functions, such as optical memory [110-111], optical transistor [112-113], optical discriminator [91, 114], optical limiter [115], optical oscillator [116-117], and optical gate [118-119].

The ideal practical bistable device for optical data processing should be small and fast, require very little switching energy and consume small amount of energy, and operate at room temperature and at a convenient wavelength. It should be able to be incorporated into an integrated system and be operable by a tiny laser diode. Recent works on the topic of optical computing using bistable property have provided new platform to envision a micrometer-size optical computing system. Lipson [120] recently demonstrates the first integrated optical system on chip. The device consists of a 5 μm-size ring resonator laser coupled with a waveguide. The entire structure is fabricated based on silicon-on-insulator (SOI) technology. The mechanism for bistability is the large thermal nonlinear coefficient in silicon which only provides 100 kHz modulation. The ring resonator provides strong light confinement that enables both the miniaturization of device dimensions and the observation of optical bistability with relatively low optical pump power [121]. Passive micron-size ring resonator has also been employed to
provide the first optical integrator [122], i.e. a capacitor-like optical device, with the processing speed of 200 GHz and holding time in order of nanoseconds.

Other devices such as FP cavity filled with nonlinear etalon can also perform fast optical computing with the appropriate materials. Nonlinear etalons can perform various optical-logic operations such as AND [123], and NOR [124] gates. The principle of such operations is changing the Fabry-Perot transmission peak by using the input pulse intensity. The nonlinear medium must be such that the absorption of one input pulse changes the refractive index at the probe wavelength enough to shift the transmission peak of the etalon by about one instrument width [125-126]. To improve the response time to picoseconds (ps) level, structure like GaAs multiple quantum well [124] can be employed as the nonlinear etalon. Measurements made by Silberberg et al. [127] suggest that the carrier lifetime can be reduced to 150 ps.

However, in general these devices have required too much optical power and were too slow to be practical. Recent discoveries have shown that the intensity levels can be reduced to 60 W/cm² for recombination times about 100 ns [128], which has allowed serious consideration for all-optical switches in telecommunication regeneration and switching systems. Moreover, with the development of optical bistable devices accessible with laser diodes, the way opened to conceive of applications in optical computing, switching and image processing. Faster devices can be accomplished by introducing surface recombination [129], proton implantation, and/or by using a second etalon in series to switch off the first [130], which enable switching speeds of 30-40 picoseconds.

Optical bistability based on FP cavity filled saturable absorption has also been studied widely for the purpose of optical switches. Most of these devices usually operate in reflection mode because it provides the highest contrast between the on and off states, since the
constructive and destructive interference due to the saturable process inside the device can create a zero-state and a maximum-state which corresponds the on and off modes. To ensure the highest contrast between the two switching states, the reflectivities of the front and back mirror $R_F$ and $R_B$ are tuned so that light reflected off the front mirror destructively interferes with light reflected back from within the cavity, which is known as impedance matching condition. This type of FP cavity is called an asymmetric FP (AFP) cavity, where $R_B$ is almost equal to 1 and $R_F$ is much smaller. An AFP, if filled with a saturable absorption or nonlinear refractive index materials, can alter the phase inside the cavity which enables reflection. This principle provides a method to modulate light optically, i.e. optically addressed spatial light modulator, through an internal field or with the assistance of electric field based on the quantum confined Stark effect (QCSE) [131].

In semiconductor quantum structures, holes and electrons occupy discrete energy levels in the spectrum. However, in the presence of an electric field, these discrete energy levels in certain materials are shifted in opposite directions. The electrons are shifted to lower energy levels while the holes move to higher energy level. This condition reduces the overlap of relating valence and conduction band in transition, which in turn reduces the possibility of transition between states. As a result, the applied electric field can be used to modulate the absorption coefficient, and consequently the refractive index.

One of the common materials used in an AFP optical switch is quantum well due to its large QCSE property. Optical switch with high contrast ratio operating on reflection mode has been fabricated in GaAs-AlGaAs multiple quantum well with 27:1 contrast ratio at 2.5 mW power [132-133], in strained InGaAs-GaAs structure with 12.2:1 contrast [134], and in GaAllnAs-AllnAs quantum well with on-off contrast ration of 1000:1 at 30 kW/cm$^2$ [135]. An alternative transmission mode for optical switch based on quantum well has been suggested to
use uniaxial thermally induced strain created externally by bonding a free standing GaAs-AlGaAs quantum well thin film to LiTaO$_3$ at high temperature, which can yield a contrast ratio of 330:1 [136]. However, works on optical computing based on simple bistable devices are difficult to achieve in large systems [137], as such the amount of research on bistable devices as two terminal system has receded in recent years.

Research done in the field of optical computing based on optical bistability in the late 80s and 90s focused much more on the studies of various modification of hybrid device such as self-electrooptict effect device (SEED) due to its simplicity in design and many other potential applications. The SEED combines a QCSE modulator and optical detection within the same structure, causing an optoelectronic feedback that, when positive, gives optical bistability. The SEED is usually described in terms of the photocurrent that the photodiode detects [138]. From the point of view of carrier transport nonlinearities, the photodiode can be considered a QCSE modulator, and the photocurrent is a manifestation of carrier transport.

A version of the SEED called the symmetric SEED (S-SEED), composing of two p-i-n quantum well devices connected in series, can act as an optical memory which includes three terminal: an individual set (S), reset (R), and complementary outputs (SR) [139]. This device allows relatively substantial processors to be constructed. A team of scientists led by Huang [140] at Bell Laboratories demonstrates a simple but complete computer [141] with four arrays of 32 S-SEED devices. This work has led to substantial efforts on architectures and on optics for handling arrays of light beams as detailed in [142]. Another team at Bell Laboratories led by Hilton constructs experimental switching machines for telecommunications routing. This work, which overlaps in time with the optical computing work, leads to several generations of experimental systems [143], including a free-space digital optical system [144] with over 60,000
light beams. This working system demonstrates the feasibility of dense free-space optical array connections.

Another variation of the SEED system is the field-effect-transistor self-electrooptic effect device (FET-SEED), which consists of doped-channel field effect transistors, multiple quantum well modulators, and p-i-n multiple quantum well detectors integrated on a single common layer substrate [145]. This device takes advantage of the spatial bandwidth available in the optical domain by integrating electronic circuits with optical detectors (input) and modulators (output) to produce what is known as smart pixel arrays, i.e. devices or units with optical inputs and outputs whose functions can be combined to enhance the complexity significantly. In the case of a FET-SEED, its logical performance is increasingly complex due to the integration of optical inputs and outputs into an electronic circuit to provide electronic gain. This approach has been successfully incorporated into a substantial system demonstration [146].

From the first experimental investigations of optical nonlinearities in semiconductors, it became clear that heating due to optical absorption could be large in wide range of materials. Because the bandgap decreases rapidly with temperature, optically induced heating will alter both the absorption and refractive index. This process represents an opto-thermal nonlinearity that has been demonstrated in a number of devices. The most important near-term application of these nonlinearities was that they enabled simple low-power observation of optical bistability in ZnSe interference filters [147], which were used as prototype nonlinear elements in studies of the potential of optical computing [148].

Because thermal heating is a nonlocal effect, it enables optical bistability without optical feedback. An absorption is required that increases with increasing intensity. This causes more absorption and the positive feedback can lead to bistability. Optothermal bistability is observed
without cavities in CdS at low temperatures [149] and also in GaAs–AlGaAs quantum wells [150], where it is shown to be a general feature of any material whose absorption is a nonlinear function of the state of material excitation. While optothermal nonlinearities can be made to occur in many semiconductor devices, their speed will be limited to thermal response times. Although heat sinking can push the speed to submillisecond, their sensitivity decreases accordingly [128]. These setbacks have limited the use of the opto-thermal bistable devices in the field of optical information processing. However, such devices can be used as forms of thermal sensor [151] by incorporating it with other optical instrument such as optical fibre, and also by using material with high sensitivity to temperature like amorphous Si.

Even though the research in optical computing in general and optical bistability in particular is no longer active today, several gains have been achieved through the process of using optical bistable devices to realize an all-optical-computing system. Two of the most notable achievements through the process of studying and developing optical bistable devices are the construction of vertical cavity surface emitting laser (VCSEL) [152-153] and optically addressed spatial light modulators (OASLM). The pioneer work on the field of VCSEL was done by Iga and his group in 1979 [154]. Jewell and colleagues later developed successfully the electrically pumped VCSEL at room temperature while working on resonant-cavity switching devices [155]. Since its commercial availability in the mid-90s, VCSEL has been used in varieties of applications ranging from low-cost optical fibre connections [156-157] to laser printing [157] and biological analysis [158].

The construction of spatial light modulator (SLM) began in the process of realization of an optical circuit system. Spatial light modulator can act as conversion of an electrical input signal to an output optical signal. This is known as electrically written SLM. Another form of
SLM, the OASLM, can input information in terms of optical signal. The OASLM has several advantages such as fast temporal response, ability to convert incoherent images to coherent images, signal amplification, and wavelength conversion. There has been more than 50 types of OASLM produced in the eighties and nineties [159]. Many different SLMs have been proposed and many prototypes fabricated, like liquid crystal SLMs [160-161], magneto-optic SLMs [162], multiple quantum wells devices [163], Si PLZT SLMs [164] and deformable mirror devices [165]. However, very few of these SLMs have survived. Today among the SLMs commercially available, mostly for display purpose, two technologies prevail: liquid crystal technology and digital micromirrors devices.

Applications of these commercially available SLMs are becoming increasingly important, due to their ability to fulfill the requirements in terms of speed, modulation capability, and resolution. The applications of SLMs are numerous, for example, recent papers have reported different application such as pulse shaping [166], quantum key distribution [167], hologram reconstruction [168], computer generated holograms [169], diffractive optical elements [170], optical tweezers [171], optical metrology [172], joint transform correlation [173], and ultrasound detection [174].

2.1.4 Models of Optical Bistability

The origin of optical bistability can be linked to the effect of either the nonlinear absorptive $\alpha$ or refractive coefficients $\gamma$, or both of these coefficients can contribute to the formation of the hysteresis. Both $\alpha$ and $\gamma$ can be described by the conventional expansion of polarization $P(t)$ in powers of the electric field $E(t)$

$$P(t) = (1)E(t) + (2)E^2(t) + (3)E^3(t) + \ldots$$

$$= P^{(1)}(t) + P^{(2)}(t) + P^{(3)}(t) \quad (2.3)$$
\(\chi^{(2)}\) and \(\chi^{(3)}\) are known as second and third order susceptibilities, respectively and \(P^{(2)}(t)\) and \(P^{(3)}(t)\) are known as the second-order and third-order polarizations, respectively. Since optical bistability is usually found in the third order nonlinearity, \(\chi^{(3)}\) is the main focus in the study. For the third order susceptibility, \(\chi^{(3)}\) can be separated into a real and imaginary parts, which can be used to identify the nonlinear absorption and refractive index. The relationships have been described by Henari et al. [175] as following

\[
\text{Re} \left( \chi^{(3)} \right) = 2n^2\varepsilon_o c \lambda y \\
\text{Im} \left( \chi^{(3)} \right) = \frac{n^2\varepsilon_o c \lambda \delta}{2\pi}
\]

where \(n\) is the linear refractive index, \(\varepsilon_o\) is the permittivity of free space, \(c\) is speed of light, and \(\lambda\) is the wavelength. For porous silicon, Henari et al.’s paper shows that \(\text{Re} \left( \chi^{(3)} \right)\) and \(\text{Im} \left( \chi^{(3)} \right)\) at 665 nm are equal to \(7.5 \times 10^{-9}\) esu and \(-1.9 \times 10^{-9}\) esu, which indicates that the change in the refractive is more dominant than the absorptive effect.

Typically most optically bistable phenomena are modelled based on the steady-state model developed first by Szoke et al. [34], and by Bonifacio and Lugiato [176-177] with mean field approximation. These models describe the FP cavity consisting of a nonlinear etalon whose effect can be either absorptive or dispersive. The first numerical study of absorptive bistability was discussed by McCall [37], and almost simultaneously Felber and Marburger [100] provided the explanation of dispersive optical bistability.

Considering a FP cavity with length \(L\) filled with a nonlinear medium as it is described in Figure 2.4. The boundary conditions of a beam of light passing through the cavity can be described as following

\[
E_B(0) = \sqrt{\varepsilon F}(0)e^{2ikL - \alpha L} \\
E_F(0) = \sqrt{\varepsilon}E_1 + \sqrt{\varepsilon}E_B(0)
\]
where $E_B$ is the backward field, $E_F$ is the forward field, $E_I$ is the input field, $E_T$ is the transmitted field and $E_R$ is the reflected field.

**Figure 2.4.** Schematic diagram of a Fabry-Perot cavity with directions of the optical fields (Adapted from [101])

Substituting (2.6) to (2.7) we can obtain

$$E_F(0) = \frac{\sqrt{TE_I}}{1 - Re^{2ikL - \alpha L}}$$

(2.12)

where $k = \frac{2\pi n}{\lambda}$ is the wave number, $R$ and $T$ are the reflection and transmission of the front and back mirrors, $\alpha$ is the absorption coefficient and $n$ is the refractive index which is taken to be real. Equation 2.12 is known as the Airy equation.

The equivalent intensity equation can easily be found by multiplying (2.12) to its complex conjugate

$$I_F = E_F(0)E_F^*(0) \equiv \frac{T_{th}}{1 + Re^{-\alpha L} [Re^{-\alpha L} - 2\cos(2kL)]}$$

(2.13)

From (2.8) we have

$$I_T = T I_F$$

(2.14)
Combining (2.13) and (2.14) we obtain the transmission characteristics of a FP cavity

\[ I_T = \frac{T^2 I_l}{1 + Re^{-\alpha L}[Re^{-\alpha L} - 2\cos(2kL)]} \]  

(2.15)

Substituting \( E_B(0) \) in (2.9) to (2.10), we can solve for \( E_R \) as a function of \( E_I \)

\[ E_B = \frac{E_R}{\sqrt{R}} \]  

(2.16)

\[ E_R = \frac{R\sqrt{Te^{2\alpha L} - \alpha L}}{1 - Re^{2\alpha L} - \alpha L} E_I \]  

(2.17)

Then the reflected intensity \( I_R \) and backward intensity \( I_B \) are

\[ I_R = \frac{R^2 Te^{-2\alpha L}}{1 + Re^{-\alpha L}[Re^{-\alpha L} - 2\cos(2kL)]} I_l \]  

(2.18)

\[ I_B = \frac{RTe^{-2\alpha L}}{1 + Re^{-\alpha L}[Re^{-\alpha L} - 2\cos(2kL)]} I_l \]  

(2.19)

Total intensity inside the cavity is the sum of the forward and backward intensity

\[ I_C = I_F + I_B \equiv \frac{T(Re^{-2\alpha L} + 1)}{1 + Re^{-\alpha L}[Re^{-\alpha L} - 2\cos(2kL)]} I_l \]  

(2.20)

If \( \alpha L \ll 1 \) and \( \cos(2kL) = 1 \), then equations (2.15), (2.18) and (2.20) become

\[ I_T = \frac{T^2 I_l}{1 + R(1-\alpha L)[R(1-\alpha L) - 2]} = \frac{T^2}{[1-R(1-\alpha L)]^2} I_l \]  

(2.21)

\[ I_C = \frac{T[1 + R(1-\alpha L)]}{[1-R(1-\alpha L)]^2} I_l \]  

(2.22)

\[ I_R = \frac{R^2 T(1-\alpha L)}{[1-R(1-\alpha L)]^2} I_l \]  

(2.23)

If we set \( C \equiv \frac{Ra}{1-R} \), also known as the cooperativity parameter, then

\[ I_T = \frac{I_l}{(1+C)^2} \]  

(2.24)

For the case of an absorptive bistability, the absorption coefficient \( \alpha \) now becomes \( \dot{\alpha} \) which can be described as a two-level saturable absorber

\[ \dot{\alpha} = \frac{\alpha_0}{1 + I_l/I_s} \]  

(2.25)
where \( I_S \) is the saturation intensity of the material and \( \alpha \) is the unsaturated absorption coefficient, and in the case of a FP cavity

\[
\dot{\alpha} = \frac{\alpha_0}{1 + I_F/I_S}
\]  

(2.26)

Combining \( \dot{\alpha} \) with \( C \), then the cooperative parameter now becomes

\[
C = \frac{R\alpha_0 L}{(1-R)(1+I_F/I_S)} = \frac{C_0}{1 + I_F/I_S}
\]  

(2.27)

where \( C_0 = \frac{R\alpha_0 L}{1-R} \)

Now the transmitted intensity of a FP cavity filled with a saturable absorber is

\[
I_T = \frac{I_I}{(1 + \frac{C_0}{1+I_{I}/I_S})^2} = \frac{I_I}{(1 + \frac{C_0}{1+I_{I}/I_S})^2}
\]  

(2.28)

Then

\[
I_I = I_T \left( 1 + \frac{C_0}{1+I_{I}/I_S} \right)^2
\]  

(2.29)

Let \( X = \frac{I_T}{I_{I/S}} \) and \( Y = \frac{I_I}{I_{I/S}} \)

\[
Y = X \left( 1 + \frac{C_0}{1+X} \right)^2
\]  

(2.30)

In order to have bistability, \( \frac{dY}{dX} < 0 \)

\[
\frac{dY}{dX} = \left[ 1 + \frac{C_0}{1+X} \right]^2 + 2X \left[ 1 + \frac{C_0}{1+X} \right] \left[ -\frac{C_0}{(1+X)^2} \right]
\]  

(2.31)

For \( \frac{dY}{dX} = 0 \)

\[
X = \frac{C_0 - 2\sqrt{C_0(C_0 - 8)}}{2}
\]  

(2.32)

This solution is only real when

\[
C_0 = \frac{R\alpha_0 L}{1-R} > 8
\]  

(2.33)
This condition limits the design of the cavity since all the parameters represent the cavity’s constant. Thus only certain types of cavity with the appropriate \( R, \alpha_o \) and \( L \) can create bistability.

For a purely dispersive bistability, we assume \( \alpha = 0 \), then the transmission of a FP cavity now becomes

\[
I_T = \frac{T^2 I_l}{1 + R[R - 2 \cos(2kL)]} = \frac{T^2 I_l}{1 + R[R - 2 \cos(2kL)\sin^2 kL]} = \frac{I_l}{1 + 4R \frac{I_l}{2} \sin^2 kL}
\]

Since this is dispersive bistability, the factor dominates the nonlinear process is the change in the refractive index. The simplest form of the nonlinear refractive index is

\[
n = n_o + \gamma I_T
\]

This composes of a linear refractive index \( n_o \) and a nonlinear refractive index \( \gamma \). Then \( I_T \) becomes

\[
\frac{I_T}{I_l} = \frac{1}{1 + 4R \frac{I_l}{2} \sin^2 \left[ \frac{2\pi L}{\lambda} (n_o + \gamma I_T) \right]}
\]

By plotting the left-hand side and right-hand side as functions of \( I_T \), the solution of the above equation represent condition for bistability at particular \( I_l \) as shown in Figure 2.2. At low intensities, the intersection point moves smoothly along the curve until it reaches B. A small change in \( I_l \) causes the point to jump to C. If \( I_l \) is decreased the intersection moves smoothly up to E where it jumps to F and a first bistable reagion is obtained. If instead, \( I_l \) is increased further from B successive bistable regions (multi-stability) can appear.

Equations (2.29) and (2.36) are the simple models for the absorptive and dispersive models of optical bistability developed by Skoze et al. A more thorough method known as the mean-field approximation method was developed by Bonifacio and Lugiato to model bistability from the Maxwell-Block equations in a two-level system.
\[
y^2 = x^2 \left[ \left( 1 + \frac{2C}{1+x^2+\Delta^2} \right)^2 + \left( \frac{2C\Delta}{1+x^2+\Delta^2} - \phi \right)^2 \right]
\]  
(2.37)

where \( y \sim \sqrt{I_i} \) and \( x \sim \sqrt{I_T} \), \( C \) is the cooperative parameter, \( \Delta \) is the phase shift, and \( \phi \) is the cavity detuning. If we set \( \Delta = 0 \) and \( \phi = 0 \), then this equation essentially becomes (2.30)

\[
y^2 = x^2 \left[ \left( 1 + \frac{2C}{1+x^2} \right)^2 \right]
\]

For purely dispersive system, \( \Delta \) is the dominant factor, i.e. \( \Delta \gg 1 \), then the dominant factor will be the second term on the right-hand side, now Bonifacio and Lugiato’s equation becomes

\[
y^2 = x^2 \left[ \left( \frac{2C\Delta}{1+x^2+\Delta^2} - \phi \right)^2 \right]
\]  
(2.38)

### 2.1.5 Optical bistability in porous silicon

PSi has been known to exhibit large third order nonlinearity, which is strongly related to optical bistability property. With the increasing interest in optical switches in the 1990s, there were several studies conducted to investigate the ability to control light with PSi material due to its simple electrochemical anodization method of fabrication at room temperature condition. Matsumoto et al. [178] first reports on the ability of large induced absorption in free standing 10-μm thick PSi layers at intensity as low as 200 kW/cm² modulated by a light chopper at 48 Hz. The sample is excited by a CW mode-locked titanium doped sapphire laser at 440 nm, and focused to a spot size of 50 μm in diameter onto the sample. The beam is then split into two strong beams acting as writing beam, and a weak beam performing the function of a reading beam, which are all focused onto the sample to test the ability of using the samples as NOR gates. However, the device only exhibits thermal process switching in the order of ms but compensated by low power requirement. The mechanism of the process is deduced to be a result of carrier localization in surface states of Si microcrystals.
After the initial result obtained by Matsumoto et al., Kanemitsu and Matsumoto [179] report on the existence of large hysteresis in the same samples at 457.5 nm. The 10-20 μm thin film is excited by using a CW Ar laser with the power up to 70 mW focused to a 50 μm spot-size. The beam is now modulated by an acousto-optic deflector at 100 Hz, which yields large hysteresis possibly due to thermal process. The physical origin of the optical bistable origin was shown to be dependent on the nonlinear absorption coefficient due to the inhomogeneities such as the nanocrystalines inside the material. The bistability curve shown in this case indicated a transient response in nature.

Study done by Maly et al. [180] also shows that PSi thin film structure can also exhibit strong bistability in the pico-second regime. However, the hysteresis curve reported also shows strong transient response. The bistability process is studied by using a Nd:YAG picosecond pulsed laser at 532 nm yielding a clockwise hysteresis cycle and fast switching time in picoseconds orders. This and Matsumoto’s results show that the induced absorption process in PSi has a fast and slow component which indicating in the study of the nature of the absorbance done by using the two beam pump-probe method. The slow component is suggested to be originated from the recombination of carriers in localized surface states, while the fast component is due to a bimolecular recombination of free carriers in the core of silicon nanocrystals.

Mesoporous PSi materials have also demonstrated the ability of exhibiting large hysteresis. Recent study done by Cojocaru et al. [28] has shown that at 593 nm, mesoporous PSi thin films can produce a pronounced hysteresis cycle in the clockwise direction with transient nature when excited with 0.9 μs pulse laser. The mechanism of the process is also explained based on the large induced nonlinear absorption. The transient hysteresis shapes
obtained by Maly et al., Kanemitsu, and Cojocaru et al. when conducting bistability study in PSi shows a strong resemblance with works done on optical bistability in amorphous silicon (a-Si). Thus, it has been suggested by Cojocaru and colleagues that mechanisms existing in a-Si like the interaction with nonequilibrium phonons and localized vibrational modes can also play an important role in the case of PSi, since this material possesses a large specific surface and, consequently, a large concentration of structural inhomogeneities of crystalline silicon at nanometric scale, that may lead to spatial localization of the phonon vibrational modes and, as a result, to the formation of nonequilibrium state for a high level of laser excitation.

Even though PSi devices have demonstrated fast switching time, this process requires switching intensity in the order of hundreds kW/cm$^2$ or even thousands of MW/cm$^2$ for picoseconds switching time. This can be improved by using a Fabry-Perot cavity instead of thin film as switching devices. Takahashi and his colleagues [29] have demonstrated the ability of switching such a device fabricated from mesoporous PSi with low intensity of 480 W/cm$^2$, and at a slow frequency of 100 Hz using an acousto-optic deflector. The hysteretic input-output curve also shows strong transient response which is modelled to be based on the effect of the nonlinear refractive index. The slow response of the device indicates the nonlinear process is entirely thermal. To explain the physical origin of the deformation of the transmitted pulse at the atomic level, Takahashi et al. uses a two-level states as a relaxation path of carriers in PSi. The strong incident optical power excites a large number of carriers to the conduction band, and then the excited carriers are thermalized to the conduction-band edge, where the thermalized carriers are accumulated due to its slow decay rate.
2. Theoretical modeling and designs

2.2.1 Fabry-Perot resonator

Recall equation (2.34) derived from section 2.1.4, which can be written as

\[ I_T = \frac{I_l}{1 + F \sin^2 \theta} \]

where \( \theta = kL \) and \( F = \frac{4R}{T^2} = \frac{4R}{(1-R)^2} \)

However, equation (2.34) ignores the existence of a linear background absorption so that

\[ E_B(0) = E_B(L) \]
\[ E_F(0) = E_F(L) \]

In many practical situations, there always exists a linear background absorption characterized by the coefficient \( \alpha \). D. A. B. Miller has shown that in this case the boundary conditions are

\[ E_B(0) = \sqrt{R} E_F(L) e^{\frac{i(kL-\alpha L)}{2}} \]  \hspace{1cm} (2.39)
\[ E_F(0) = \sqrt{T} E_I + \sqrt{R} E_B(0) \]  \hspace{1cm} (2.40)
\[ E_T = \sqrt{T} E_F(L) \]  \hspace{1cm} (2.41)
\[ E_R = \sqrt{R} E_B(0) \]  \hspace{1cm} (2.42)
\[ E_F(L) = E_F(0)e^{\frac{i(kL-\alpha L)}{2}} \]  \hspace{1cm} (2.43)

Then, the transmission now becomes

\[ \tau = \frac{I_T}{I_I} = \frac{T^2 e^{-\alpha d}}{(1-Re^{-\alpha d})^2} \frac{1}{1+F \alpha \sin^2 \theta} = \frac{(1-R)^2 e^{-\alpha d}}{(1-Re^{-\alpha d})^2} \frac{1}{1+F \alpha \sin^2 \left( \frac{2\pi nd}{\lambda} \right)} \]  \hspace{1cm} (2.44)

where \( F \alpha = \frac{4Re^{-\alpha d}}{(1-Re^{-\alpha d})^2} \), and \( \theta = \frac{2\pi nd}{\lambda} \)

Using the boundary conditions (2.39)-(2.43) and equation (2.44), it is noted that the intensity inside the cavity is significantly enhanced compared to the input intensity, and also is
much larger than the transmitted intensity. In a case of perfect transmission, the intracavity intensity is enhanced by a factor of \( \frac{1+R}{1-R} \), which can be very large if the mirrors are highly reflective.

Equation (2.44) shows that the FP cavity depends on constant parameters like the mirror transmission and reflectivity, material background absorption coefficient. To understand the effect of the cavity mirrors reflectivities, a simulation of the transmission and the intra-cavity intensity at different mirrors reflectivities were conducted as detailed in Figure 2.5. The intra-cavity intensity shows a dramatic increase as the cavity mirrors become reflective which can be utilized to lower the input intensity.

**Figure 2.5.** Variations in the transmission (left) and the intracavity intensity (right) of a Fabry Perot cavity for different values of reflectivities. The simulations were done using the same input intensity.

The transmission also improves with higher reflectivities, and becomes very sharp peaks centered about integer multiples of \( \pi \). To describe the quality a FP mirror, two parameters known as the finesse and the Q-factor are usually utilized. The finesse is related directly to the mirrors reflectivities as following

\[
F = \frac{\pi \sqrt{F_\alpha}}{2}
\]  

(2.45)
And the quality factor can be defined through its relationship with $F$ via definition

$$Q = \frac{\lambda_o}{\Delta \lambda} = \frac{\lambda_o}{\lambda_f} F$$

(2.46)

where $\lambda_o$ is the resonant wavelength, $\Delta \lambda$ is the linewidth, $\lambda_f$ is the separation wavelength between two transmission modes. $\frac{\lambda_o}{\lambda_f}$ is the phase response of the cavity and can be determined as

$$\frac{\lambda_o}{\lambda_f} = \frac{2\pi d}{\lambda_o}$$

(2.47)

where $n$ is the refractive index of the material that make up the cavity, and $d$ is the cavity length.

As a result, if the reflectivities of the mirrors are increased, the finesse and the Q-factor also increase. In other words the higher the Q-factor, the higher the intra-cavity intensity which is beneficial to the bistability process. Another parameter that can influence the performance of the cavity is the material absorption coefficient. It is easy to show that increasing the absorption inside the cavity will reduce the transmission as well as the intracavity intensity due to the fact that the factor $\frac{T^2 e^{-\alpha d}}{(1 - Re^{-\alpha d})^2}$ decreases as $\alpha$ gets larger.

The Q-factor can also tuned by making the resonant wavelength to be at higher wavelength or by increasing the cavity length. However, there are certain limits with these methods because by making the cavity longer, this also increases background material which also increases the absorption coefficient. Moreover, even though it is desirable to have a cavity operating in the long wavelength region, this region also has a limited potential excitation light source due to the limitation of technology.
2.2.2 Fabry-Perot cavity filled with nonlinear refractive index material

As described above, equation (2.42) represents a FP with linear refractive index material etalon. It can be rewritten by introducing two new parametric variables \( X \) and \( Y \) which are defined as

\[
X = \left( \frac{2\pi d y}{\lambda} \right)^3 \frac{(1-R)(1+Re^{-\alpha d})(1-e^{-\alpha d})}{\alpha d(1-Re^{-ad})^2} I_l
\]

(2.48)

\[
Y = \left( \frac{2\pi d y}{\lambda} \right)^3 \frac{(1+Re^{-\alpha d})(1-e^{-\alpha d})}{\alpha d(1-R)e^{-\alpha d}} I_T
\]

(2.49)

where \( d \) is the cavity length, \( \lambda \) is the excited wavelength, \( R \) is the mirror reflectivity, \( \alpha \) is the linear absorption coefficient, and \( \gamma \) is the nonlinear refractive index.

\( X \) and \( Y \) are then related to each other as following

\[
X = Y \left[ 1 + F_\alpha \sin^2 \left( \frac{1}{3} \gamma \delta_\alpha \right) \right]
\]

(2.50)

where \( F_\alpha = \frac{4Re^{-\alpha d}}{(1-Re^{-\alpha d})^2} \), and \( \delta_\alpha \) is the phase detuning.

Equations (2.48), (2.49) and (2.50) describe the characteristic performance of a Fabry-Perot cavity filled with a nonlinear refractive index material. It was first used to describe the bistability behavior of a FP cavity filled with CdHgTe by Xiaoguang et al [181]. These relationships are similar to equation (2.44). If we divide (2.48) by (2.49) then we have,

\[
\frac{X}{Y} = \frac{(1 - R)^2 e^{-\alpha d}}{(1 - Re^{-ad})^2} \frac{I_l}{I_T} = 1 + F_\alpha \sin^2 \left( \frac{3}{\sqrt{Y}} \right)
\]

or

\[
\frac{I_T}{I_l} = \frac{\frac{\gamma^2 e^{-\alpha d}}{(1-Re^{-ad})^2} \frac{1}{1+F_\alpha \sin^2 \left( \frac{3}{\sqrt{Y}} \right)}}
\]

(2.51)

which is similar to (2.44) if the nonlinear phase change \( \frac{3}{\sqrt{Y}} \) is replaced by a linear process.
The nonlinear coefficient $\gamma$ is described by using Takahashi et al.’s model [29], when they explain the changes in the refractive index of a PS micro-cavity as a function of excited intensity in the 800 nm region.

$$\gamma = \beta \int_0^t l_{\text{out}}(t') e^{-\mu t'} dt'$$

(2.52)

where $\beta$ and $\mu$ are the two fitting parameters are used to describe the changes in the nonlinear refractive index $\gamma$. $\beta$ is called the nonlinear coefficient and $\mu$ is the transient frequency, so $\tau = \frac{1}{\mu}$ is the transient lifetime of the bistability process.
III. EXPERIMENTAL PROCEDURES

1. Porous silicon fabrication and surface passivation

PSi is typically fabricated by electrochemically etching crystalline Si under constant current conditions in an electrolyte containing hydrofluoric acid. The process leads to the formation of thin films of crystalline silicon permeated with a matrix of voids of well defined size and porosity. The pore sizes and morphology can vary significantly depending on the electrochemical reaction conditions \[17\]. For optical wavelengths the films appear as a uniform dielectric material with the refractive index linked to porosity using a mean effective medium model. When the etching current is changed during the fabrication the PSi films will adopt a different porosity, leaving the preexisting films unchanged. This property allows multilayered structures with regions of different porosity to be formed as a single monolithic film \[182\].

In this study PSi photonic structures are formed by anodic etching of heavily boron doped (Resistivity = 1.5 - 2 mΩ.cm), <100> oriented Si wafers in an ethanolic hydrofluoric acid (25%) solution at room temperature. These conditions lead to the formation of mesoporous type PSi with pore sizes in the range of 10 to 100 nm; the porosity and film thickness may be controlled by appropriate selection of applied etching parameters. Optically bistable devices were composed of two dielectric mirrors bounding a half-wavelength thick cavity layer; the mirror regions consisted of five bi-layer periods of alternately high and low porosity. For the purposes of conducting transmission measurement the PSi films were removed from its native Si substrate using an electropolishing step using a high current pulse in a lower concentration (15%) hydrofluoric acid solution. The free standing films are then transferred to a transparent glass substrate.
The pore formation can be characterized by the following general anodic reactions

\[ \text{Si} + 6\text{HF} \rightarrow \text{H}_2\text{SiF}_6 + \text{H}_2 + 2\text{H}^+ + 2e^- \]

And the electropolishing process is

\[ \text{Si} + 6\text{HF} \rightarrow \text{H}_2\text{SiF}_6 + \text{H}_2 + 4\text{H}^+ + 4e^- \]

In order to investigate the influence of the porosity of the central layer, optical bistable devices were fabricated with different porosities in the central cavity layer ranging from medium (60%) to high (90%) porosity; the exact etching parameters for each structure are provided in Table 1. A detailed set up of the anodization process is shown in Figure 3.1 (A) and 3.1 (B). The etching process occurs inside the electrochemical cell. The cell consists of a stainless steel electrode used for mounting the silicon wafer. It is sealed through a 1.0 cm diameter Viton o-ring, so that only the polished surface of the wafer is exposed to the electrolyte. In the cell, a platinum electrode submerged in the solution acts as the cathode, while the silicon wafer performs the function of an anode. The electrolyte temperature is monitored using a Teflon coated thermocouple. The power to the cell is supplied by a power source operated in constant current mode controlled via a computer running LabView program. The cell is connected in series with a 1 k\(\Omega\) resistor to ensure a more stable current. The current is monitored through an oscilloscope to ensure stable operating conditions.
Figure 3.1. Experimental set up of the etching process. (A) Actual configuration of the electrochemical cell under a fume cover. (B) Circuit used for the anodization process.

As this study is concerned with porous silicon, the effective medium approximation derived by Looyenga was employed to link the refractive index of PSi to the structure porosities and the refractive index of silicon and air. This model has been verified to be topologically more appropriate for mesoporous materials than the other theories [183-184]. The effective refractive index has the form [185]

\[ n_{PSi} = \sqrt[3/2]{(1-p)n_{Si}^{2/3} + pn_{air}^{2/3}} \]  

(3.1)

where \( p \) is the porosity of the layer, \( n_{Si} \) and \( n_{air} \) are the refractive indices of crystalline silicon and air.
For temperatures between $0^\circ C$ and $500 ^\circ C$ and in the energy region from $1 \text{ eV}$ to $3.648 \text{ eV}$, the refractive index of crystalline silicon has been shown to satisfy the expression shown in literature [186]

\[
n(E, T) = n_0(E) + a(E)T
\]  

(3.2)

where $T$ is the temperature expressed in C, and in function of the energy (eV):

\[
n_0(E) = \left[ 4.565 + \frac{97.3}{(E_g^2 - E^2)} \right]^{1/2}
\]  

(3.3)

\[
a(E) = \left[ -1.864 + \frac{53.94}{(E_g^2 - E^2)} \right] \times 10^{-4}
\]  

(3.4)

where $E_g$ is the direct bandgap of crystalline Si and has the value of $3.648 \text{ eV}$.

**Table I.** Etching conditions

<table>
<thead>
<tr>
<th>Porosities (%)</th>
<th>Layers</th>
<th>Current (A)</th>
<th>Etch time (s)</th>
<th>Structure</th>
</tr>
</thead>
<tbody>
<tr>
<td>60%</td>
<td>Low</td>
<td>0.005</td>
<td>14.58</td>
<td>L(HL)$_4$A$_2$(LH)$_5$</td>
</tr>
<tr>
<td></td>
<td>High</td>
<td>0.22</td>
<td>1.06</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Active</td>
<td>0.02</td>
<td>6.00</td>
<td></td>
</tr>
<tr>
<td>71%</td>
<td>Low</td>
<td>0.005</td>
<td>14.58</td>
<td>L(HL)$_4$A$_2$(LH)$_5$</td>
</tr>
<tr>
<td></td>
<td>High</td>
<td>0.22</td>
<td>1.06</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Active</td>
<td>0.05</td>
<td>3.10</td>
<td></td>
</tr>
<tr>
<td>80%</td>
<td>Low</td>
<td>0.005</td>
<td>14.58</td>
<td>L(HL)$_4$A$_2$(LH)$_5$</td>
</tr>
<tr>
<td></td>
<td>High</td>
<td>0.22</td>
<td>1.06</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Active</td>
<td>0.22</td>
<td>1.06</td>
<td></td>
</tr>
<tr>
<td>90%</td>
<td>Low</td>
<td>0.005</td>
<td>14.65</td>
<td>L(HL)$_4$A$_2$(LH)$_5$</td>
</tr>
<tr>
<td></td>
<td>High</td>
<td>0.22</td>
<td>1.06</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Active</td>
<td>0.3</td>
<td>0.85</td>
<td></td>
</tr>
</tbody>
</table>
In addition to porosity, we also investigate the role of the surface chemistry on the switching properties by modifying the silicon surface with neat undecylenic acid through a hydrosilylation process at 120°C under an Argon environment [187]. The resulting Si-C bonds are highly stable and inhibit the formation of native oxide, which readily forms on the hydrogen terminated silicon surfaces which are present directly after etching. Hydrosilylation chemistry has previously been shown to be effective for controlling quenching effects in luminescent PSi [188].

Figure 3.2 shows a detailed set up of the hydrosilylation method. The actual experimental process was conducted in the following way. The porous silicon lift-off films were placed in the dry and argon-filled reaction vessel (a custom-made Schlenk flask), with a degassed (through a minimum of 5 freeze–pump–thaw cycles) sample of neat undecylenic acid. The sample was kept under a stream of argon while the reaction vessel was immersed in an oil bath set to 120 °C for 16 hours. The flask was then opened to the atmosphere, and the functionalized surface sample rinsed consecutively with copious amounts dichloromethane and ethanol and then blown dry under a gentle stream of argon, before being either analyzed or further reactions.

<table>
<thead>
<tr>
<th>Passivation Level</th>
<th>Treatment</th>
<th>L(HL)_4A_2(LH)_5</th>
</tr>
</thead>
<tbody>
<tr>
<td>71% Low</td>
<td>0.005</td>
<td>14.58</td>
</tr>
<tr>
<td>71% High</td>
<td>0.22</td>
<td>1.06</td>
</tr>
<tr>
<td>71% Active</td>
<td>0.05</td>
<td>3.10</td>
</tr>
<tr>
<td>80% Low</td>
<td>0.005</td>
<td>14.58</td>
</tr>
<tr>
<td>80% High</td>
<td>0.22</td>
<td>1.06</td>
</tr>
<tr>
<td>80% Active</td>
<td>0.22</td>
<td>1.06</td>
</tr>
<tr>
<td>90% Low</td>
<td>0.005</td>
<td>14.58</td>
</tr>
<tr>
<td>90% High</td>
<td>0.22</td>
<td>1.06</td>
</tr>
<tr>
<td>90% Active</td>
<td>0.3</td>
<td>0.85</td>
</tr>
</tbody>
</table>
2. Optical parameters characterization

In order to understand the behavior of the bistable hysteresis, a number of optical parameters need to be known to successfully model the nonlinear input-output relationships. These parameters are the reflectivities of the mirrors and the effective absorption of the devices. The method used is called the spatially resolved reflectivity to measure the narrow spectrum of the resonance cavity. Since the samples’ surfaces are not entirely homogeneous, this can result in an increase in the FWHM of the resonance. To avoid the effect of these inhomogeneities, small spot sizes were used to probe the cavities. Another advantage of this method is the ability to probe the samples at various positions on their surfaces.

Figure 3.3 outlines the spatially resolved reflectivity set-up is used to measure the resonance spectrum of various samples. Normal incidence white light reflectivity and transmission measurements were used to characterize the optical properties of the bistable devices. White light was coupled to a single mode fiber, which then passed through a series of lenses used to control the spot-size. Light was collimated after passing through the first lens, and
then a variable aperture, and finally focused onto the sample by a 20cm focal length lens. After focusing the beam onto the sample, the reflected light was collected by using a fiber coupler connected to a compact spectrometer (Ocean Optics USB2000), by deflecting using a non-polarizing cubic beam splitter. The sample was mounted on a XYZ translational stage which can be used to measure the trace the resonant modes at different positions of the same sample.

![Image](image.jpg)

**Figure 3.3.** Spatially resolved measurement used to characterize the cavities.

To measure the transmission characteristics of the cavities, the XYZ stage positioned such that the collimated beam passed through the sample and then reflected from a mirror positioned 45, so the transmitted beam was parallel to the bench. The transmitted light was collected by the optical coupler connected the Ocean Optics spectrometer. The data collected from the spectrometer was analyzed by a LabView program, and then utilized the data to plot the reflection/transmission spectrum. The program functioned by obtaining the information of the background light \(B\), the reference beam \(R\) and the transmitted \(Tr\) or reflected beam \(Re\). The spectrum \(S\) is calculated by \(S = \frac{Tr-B}{R-B}\) for the case of transmission and as \(S = \frac{Re-B}{R-B}\) for the reflection measurement.
3. **Optical bistability measurement**

Bistability measurements were performed using a frequency doubled, Q-switched 532 nm Nd:YAG (Quantronix) laser with a pulse width 150 ns, repetition rate of 1 kHz, and average power in the range of 0.5 to 21 mW. The experimental arrangement is depicted in Figure 3.4(A): the incident beam is focused onto the sample using a 60 cm focal length lens, creating a spot size of approximately 90 μm in diameter on the sample. A partial mirror is used to deflect a small fraction of the beam to a 1 GHz fast photodiode (Hamamatsu S9055) which was used to monitor the input intensity. The samples were mounted on a rotational stage for the purpose of changing the incident angle. The transmitted light was measured as a function of incident angle and was then fixed at an angle corresponding to the cavity resonance. The transmitted light was collected with a second fast photodiode with matched response times. The photodiode detectors are mounted on XYZ translational stage for fine adjustment. Both input and output pulses were then collected using a digital oscilloscope (Tektronix TDS 2004B, 60 MHz). Absolute powers were measured using a calibrated photodetector (Coherent Inc. Field Master).

To test the thermal effect of PSi samples, the Q-switched laser was replaced by a small (65 mm in length and 11 mm in diameter) 10 mW continuous-wave (CW) diode laser (Jaycar) operating at 532 nm. The diode laser was powered by a power supply manufactured by Opto Power Corporation (OPC-PS03-A). The operating current was from 150 – 300 mA, which provided a maximum output power of around 10mW. The optimal working current was found to be in the range of 130 - 230 mA. The power supply automatically switched off when the current increased closed to 300 mA to protect the laser from overload.
The focused spot-size of the diode laser onto the sample was 0.53 mm by using the 60 mm focus length. The beam was modulated by an optical chopper (Standford Research System-SR540) connected to a controller which can increase the frequency up to 3.7 kHz. The experiment was conducted at 230 Hz and then gradually increased to 3.1 kHz. The two fast detectors were replaced by two slower silicon detectors to detect the pulses. A square wave was used as the input to the power supply through a function generator. The shape of the square wave function was optimized by including an offset voltage from the function generator. The information of the input and transmitted pulses collected by the digital oscilloscope in all experiments was obtained by the program OpenChoice Desktop Application, which was then used to plot the hysteresis curves. The schematic diagram of the thermal setup is shown in Figure 3.4 (B).

**Figure 3.4.** Optical bistability measurement for (A) fast process by using a Nd:YAG pulsed laser and (B) thermal process using a diode laser modulated by an optical chopper.
IV. RESULTS

1. Transmission/reflection spectrum studies

Figure 4.1 (A-D) shows the reflection and transmission spectrum of unpassivated the bistable devices under low intensity illumination condition. The transmission spectra at longer wavelengths are slightly greater than 1 which can be due to misalignment resulting in lowering the reference values. At normal incidence the resonant modes are positioned at 563.84 nm, 567.55 nm, 563.87 nm and 575.02 nm. From the reflection and transmission of the spectra of all the samples it is noted that the overall loss in the cavity due to absorption and scattering are approximately 20%, indicating by the gap between the transmission and reflection resonant peaks. However, it is also observed that the losses decrease as the porosities of the samples increase which can indicate increasing trend of the Q-factors. The linewidths of the cavities spectra for these samples are measured to be 7.18 nm, 6.01 nm, 5.60 nm and 5.59 nm for porosities ranging from 60% to 90% by fitting the transmission resonance to the Lorentz functions shown in Figure 4.2 (A-D). The Q factors for the individual cavities are determined by equation (2.46) which yields results of 78.5, 94.4, 100.7 and 102.9. These numbers show an improvement in the quality of the cavity with increasing porosities, which can be explained in terms of the decreasing absorption coefficient of the samples as the materials become more porous. As the porosities increase, there is less silicon existing in the device, which reduce the absorption effect and thus improving the finesse of the cavities.
Figure 4.1. Reflection and Transmission spectrum of the empty cavities with porosities ranging from (A) 60%, (B) 71%, (C) 80%, to (D) 90%.

Figure 4.2. Lorentzian fits of the transmission resonance of the unpassivated samples with porosity of (A) 60%, (B) 71%, (C) 80%, and (D) 90%.
A central effect of the surface passivation process is the red-shift of the resonant mode after being passivated, as shown in Figure 4.3. However, to make valid comparison between different cavities, the resonant modes of all the samples should be fairly consistent in terms if their widths and the peak positions. Thus, the cavities used for passivation were fabricated in such a way that after the hydrosilylation process, the new cavity resonance should be similar to the unpassivated ones with the same porosities. For the passivated samples used in this study, their resonant peaks are observed to be at 561.12 nm, 592.58 nm and 584.21 nm as shown in Figure 4.4 (A-C) for porosities of 71%, 80% and 90% respectively. The Lorentzian fit of the cavity resonance indicates the FWHM of resonant spectra to be 6.63 nm, 6.15 nm and 5.95 nm for samples with porosities ranging from 71% to 90%, which indicate Q-factors of 90.46, 96.35 and 98.19. Overall, the FWHM of the passivated samples’ resonant spectra are quite similar to the resonant width of the unpassivated devices with the same porosities. The resonant peaks positions are also quite closed to the peaks of the unpassivated samples. These features indicate a homogenous characteristic of the cavities and provide a standardized basis to compare the bistable behaviors of different cavities.

**Figure 4.3.** Shift in the resonant mode after the sample is treated with hydrosilylation chemistry. An observation of almost 100nm red-shift is observed. An increasing in transmission is possibly due to the fact that the mode is in a less absorbing region.
In order to tune the resonance to the excitation wavelength the angle of incidence was adjusted so that the cavities can couple their resonant modes to the laser line at 532 nm since the resonant peaks are centered at wavelengths well above 532 nm. Figure 4.5 shows the transmission versus angle of incidence for a 71% unpassivated structure. The experiment was conducted by rotating the samples and measuring the transmission at particular incident angles with the 532 nm Nd-YAG laser and the transmitted power was monitored via the oscilloscope. The samples were positioned at the optical axis so that when they rotated, the incident laser spot on the sample stayed consistent. It can be easily observed from Figure 4.5 that the resonant position of the cavity is shifted to shorter wavelengths (blue-shift) when the incident angle increases. This behavior has been noted in literature [189-190] for multilayered structures composed of quarter-wave thick Bragg reflectors. There also exists an intrinsic relationship between the effective refractive index $n_{eff}$ of the active region of the structure, the incident angle rotation $\theta$ and the original wavelength $\lambda_0$ as following $\lambda_\theta = \lambda_0 \sqrt{1 - \left(\frac{\sin \theta}{n_{eff}}\right)^2}$ [189]. Another important feature that
should be considered is that when the incident angle becomes very large, this can induce an optical loss and leads to the broadening of the linewidth.

\[\text{Figure 4.5. Typical angle dependent transmission of mesoporous silicon microcavities.}\]

2. Bistability experiments

4.2.1 Empty Cavities

a) Pulse deformation and hysteresis studies

For switching experiments we find that above a given threshold intensity the transmitted pulse shape changes with respect to the input pulse. The cavities were rotated at angles of 49°, 42°, 41°, 39°, 49°, 42° and 41° for 60%, 71%, 80%, 90% and passivated 71%, 80% and 90% samples respectively to couple to the laser line at 532 nm. The nature of the deformation is dependent on (i) the peak intensity, (ii) the porosity of the central layer, and (iii) the surface passivation. Figure 4.6 shows the pulse deformation after passing through the cavities, and Figure 4.7 shows the corresponding optical hysteresis of different samples, plotted as the output intensities as a
function of the input intensities. The hysteresis curves have a shape typical of transient effects as mentioned in previous study [105].

![Hysteresis Curve](image_url)

**Figure 4.6.** Pulse deformation after the input pulse passes through the sample

Table II summarizes the onset and damage threshold as a function of porosities. For the central layer with porosity of 60%, the sample starts to show pulse deformation at 846 kW/cm², observed as an obvious hysteresis in the clockwise direction as shown in Figure 4.7 (left). The FWHM of the transmitted pulse becomes much smaller with increasing input intensity, consequently increasing the area of the hysteresis curve. When the intensity becomes larger than 1038 kW/cm², the laser starts to damage the sample.

When porosity is increased to 71%, the hysteresis is observed at lower intensity of 769 kW/cm² which corresponds to a large pulse deformation. The hysteresis becomes larger with increasing input intensities, as in the case of the 71% porosity sample, but becomes smaller at very high intensity of 1115 kW/cm² which indicates that the sample is damaged. The damage is also confirmed by the presence of observable marking on the device. Similarly, the 80% porosity sample exhibits onset intensity at 653 kW/cm², and damaged threshold at 1268 kW/cm²; while
the very high porosity of 90%, its onset intensity is at 538 kW/cm\(^2\) and its damaged threshold is 1553 kW/cm\(^2\).

**Figure 4.7.** Optical hysteresis at various input peak intensities conducted in (left) transmission mode, and (right) in transmission mode.

<table>
<thead>
<tr>
<th>Porosities (%)</th>
<th>Onset intensities (kW/cm(^2))</th>
<th>Damaged thresholds (kW/cm(^2))</th>
</tr>
</thead>
<tbody>
<tr>
<td>60%</td>
<td>846</td>
<td>1038</td>
</tr>
<tr>
<td>71%</td>
<td>769</td>
<td>1115</td>
</tr>
<tr>
<td>80%</td>
<td>653</td>
<td>1268</td>
</tr>
<tr>
<td>90%</td>
<td>538</td>
<td>1553</td>
</tr>
<tr>
<td>71% (passivated)</td>
<td>1334</td>
<td>1830</td>
</tr>
<tr>
<td>80% (passivated)</td>
<td>1151</td>
<td>1946</td>
</tr>
<tr>
<td>90% passivated</td>
<td>899</td>
<td>2197</td>
</tr>
</tbody>
</table>

Strong bistability is also observed when the experiment is conducted in reflection; in this case the hysteresis moves in a counterclockwise direction as demonstrated in Figure 4.7 (right).
The input-output relationship also shows strong transient response, and the areas of the hysteresis are similar to those conducted in transmission mode in the same ranges of excited intensities for the same sample. The counterclockwise direction indicates that the reflected pulse becomes wider when it is reflected from the sample. This behavior is opposite to a steady state bistable process.

For a normal optical bistability process conducted in reflection mode, the reflected intensity increases linearly until the input intensity reaches certain value, then the output dramatically decreases. If the input intensities start to decrease, this will result in a dramatic increase of $I_{output}$ at certain excitation value. In other words, the hysteresis curve can be traced from the upper branch till it reaches the switching value, then it will switch to the lower branch, and will switch back to the upper branch if the input decreases to a critical value. This process results in a reflected hysteresis moving in a clockwise direction. A similar process happens in the case of the transmitted bistability, but now the hysteresis moves in the counterclockwise direction.

Samples which have been passivated with the hydrosilylation chemistry also exhibit bistability with transient hysteresis, however the process is now observed at intensities higher than their unpassivated counterparts as shown in Table 2. The damaging threshold of the passivated samples is also observed to be higher than the unpassivated samples with the same porosities, which shows an improvement in the structures tolerance. The onset intensities for samples with porosities of 71%, 80% and 90%, after being treated with surface passivation process, are now shown to be at 1334 kW/cm$^2$, 1151 kW/cm$^2$ and 899 kW/cm$^2$ respectively. The damaged thresholds are now equal to 1830 kW/cm$^2$, 1946 kW/cm$^2$, and 2197 kW/cm$^2$. These values also indicate a similar trend of porosity dependence for the bistability process as those
unpassivated samples. The switching intensities decreases as the porosity increases while the
damaged threshold increases with higher porosities.

Two low porosities of 47% are also studied for optical bistable properties, one was
treated with hydrosilylation chemistry and the other sample was not. These samples composed of
two six-layers Bragg mirrors instead of five-layers with the porosity of the active region was
47%. However they did not show any bistability, and the samples damaged threshold were
recorded to be 284 kW/cm$^2$ for the unpassivated cavity and 393 kW/cm$^2$ with the passivated
sample. This result also agrees with the observation found in the studies done with five-periods
mirrors in terms of the improvement in the structure toughness when treated with surface
passivation.

b) Intensity dependent transmission study

To understand the dynamics of the bistability process, an intensity dependent study is undertaken
for all the samples by plotting the transmission (input/output) as a function of time. Figure 4.8
shows this nonlinear transmission process when a high power laser pulse passes through the
sample. When the intensity starts to increase, the transmission process remains rather constant,
but when the input reaches certain intensity, the transmitted intensity begins to decrease which
Corresponds to a nonlinear change in the refractive index of the cavity. This effect continues until
the input intensity is lowered to a certain value and the nonlinear change becomes weaker, which
allows a linear transmission process. As the peak pulse intensity is increased, a stronger decrease
in the transmission takes place, this process corresponds to an increase in the hysteresis curve as
shown in Figure 4.7 (left). At very high intensity, all the samples exhibit a quite linear
transmission process due to the fact that the sample is damaged. The dip in the transmission
process takes place in a period of approximately 100 nS, and the change in the transmission process can be as high as 80%, indicating a large pulse deformation at very high intensity.

Another way to understand the nonlinear transmission process is through understanding the resonant mode of the cavity. As it has been shown in previous studies [33, 191], porous silicon microcavities are very sensitive to change in refractive index corresponding to a shift in the resonant mode position. If the input intensity of the excited laser pulse is large enough to induce a significant increase in the cavity's refractive index, this will cause a red-shift in the resonant cavity which will make the cavity become uncoupled from the laser beam. This corresponds to a drop in the transmission. When the input intensity starts to decrease, this induces a blue-shift in the cavity mode, so the resonant mode moves back to the original position, i.e. the material becomes more transparent. If the intensity is small enough, a linear transmission can be observed again which corresponds to the horizontal parts of the curves in Figure 4.8.

![Figure 4.8](image)

**Figure 4.8.** Intensity dependent transmission process.
4.2.2 PSi Cavity Doped with Colloidal Quantum Dots (Q-dots)

The sample was fabricated by depositing a 565 nm colloidal quantum dots onto a six-period Bragg reflector, and then a lift-off six-period Bragg mirror was placed on top of the Q-dots and the other mirror. The Q-dots used in the experiment are commercial colloidal quantum dots called Invitrogen-QDOT Streptavidin Conjugate consisting of a cadmium selenide/zinc sulfide CdSe/ZnS compound semiconductor core-shell heterostructure surrounded by a polymer coating and conjugated with streptavidin molecules on the outer surface with the emission band at 565 nm. The sample was assembled on a glass substrate of the purpose of transmission study. Such a structure has shown to enhance the luminescence of the cavity by ten times, and the linewidth of the resonant cavity is consistent with an undoped structure of the same porosity effective layer [23].

The sample was first studied by using the Nd:YAG pulsed laser for optical bistable property. The observed hysteresis is recorded and plotted in Figure 4.9. From Figure 4.9, it is observed that the switching intensity is at 34 kW/cm$^2$ and the damaged threshold is around 384 kW/cm$^2$. The hysteresis curve expands its area from 34 kW/cm$^2$ to 346 kW/cm$^2$, and starts to decrease at around 384 kW/cm$^2$ which indicates that the structure has been damaged at high intensity. The bistable curve still shows a pronounced clockwise-direction hysteresis as observed in the case of the empty cavity structures. The transmitted pulse linewidth is recorded to be smaller compared to the input pulse expressing similar ability of pulse shaping as in the previous studies with empty cavities. The intensity dependent nonlinear transmission plot as functions of time is shown in Figure 4.10. The transmission is constant at low intensity, as the peak intensities increase, the transmission starts to decrease indicating the beginning of the nonlinear process.
The time scale for the nonlinear transmission process is recorded to be at 100 nS before the transmission curve becomes plateau.

**Figure 4.9.** Optical hysteresis as function of peak intensities when excited with Nd:YAG pulsed laser. At 384 kW/cm$^2$, the sample is damaged so that the hysteresis curve becomes smaller even though it is excited at very high intensity.

**Figure 4.10.** Intensity dependent transmission of the quantum dot doped cavity.
To study the thermal response of the sample, the pulse laser was replaced by the CW diode laser and the beam was modulated by an optical chopper. At 230 Hz, the sample shows strong bistable property which implies large deformation of the square-wave like pulse after passing through the sample at 0.90 W/cm², corresponding to a power of 4mW from the diode laser. The input intensity increases from 28 mW/cm² to 4.05 W/cm² creating a series of hysteresis shown in Figure 4.11. The hysteresis was plot as functions of the logarithms of the measured voltages from the input and output. The curves again rotate in a clockwise direction. At low intensity of 28 mW/cm², the response of the sample is linear which represents as a straight line. However, when the laser power is turned up which produces subsequent intensities of 899 mW/cm², 1.80 W/cm² and 4.05 W/cm², the sample starts to exhibit nonlinear response like bistability, which expresses through a series of hysteresis.

![Figure 4.11](image)

**Figure 4.11.** Optical hysteresis when excited with CW diode laser at different intensities.

Hysteresis shown in Figure 4.11 is the result of the pulse deformation at high intensity as observed in Figure 4.12. The input is a square-wave like pulse with curly rising and relaxing parts and the changes in the pulse shapes occur at these positions. From Figure 4.12, if we trace...
the pulse shape as function of time, it is easily observed that from 0 ms to 0.5 ms the input and output pulses overlap each other. As time increases, the deformation starts to happen. The transmitted pulse now is now rising faster the input pulse, i.e. at a certain time instance the output is recorded to be at higher value than the input. This discrepancy between the input and the transmitted pulses create the hysteresis. When the intensity reaches its maximum amplitude, the pulse deformation is no longer observed which corresponds to the linear parts in the hysteresis. This process repeats itself as another pulse in the long pulse train passing through the sample.

**Figure 4.12.** Pulse deformation at 230 Hz. The transmitted pulse has been expanded rather than contracted as when it is excited by the Nd:YAG laser.

Figure 4.13 shows the intensity dependent inverse transmission by plotting the ratio of input/output as function of temporal response. Since the output is larger than the input, an inverse of the transmission is taken so that the curves still follow the same pattern as when the excited source is the Nd:YAG laser shown in Figure 4.10. Overall, the changes in the transmission response are intensity dependent and become larger with increasing excitation.
power. When the response is linear, the transmission is presented as a constant horizontal line, and as the nonlinear process starts to happen, a sharp decrease in the inverse transmission curve is observed. However, these changes start to recover themselves and become linear again. This corresponds to the process when the input and output pulse overlap each other at maximum amplitude.

![Graph showing intensity dependent transmission of the Q-dot doped cavity](image)

**Figure 4.13.** Intensity dependent transmission of the Q-dot doped cavity when excited by the diode laser and modulated by the chopper.

The hysteresis shown in Figure 4.14 becomes smaller and the response is almost linear when the frequency response becomes faster. The plot compares the hysteresis responses at 230 Hz and 3.1 kHz. At 3.1 kHz the hysteresis almost disappears, which confirms the process observed is thermal since thermal response is usually quite slow and in the in order of a few hundred milliseconds. In general, this information indicates that the sample has both the thermal and fast switching properties which can be due to the role of the quantum dots. Quantum dots have been shown to be able to have bistable properties in milliseconds to microseconds time scale with intensity as low as 250 W/cm$^2$. This means that the thermal response can be due to the response of the quantum dots. However, when a study of bistability in quantum dots was
conducted with the 550 nm QDot solution in a cuvette and an interference filter at 530 nm using the diode laser and the pulsed laser, no bistability was observed. An empty cavity with the same porosity was also tested for bistability using the diode laser, and it also yields no result. As a result, it is inconclusive that which mechanism is the origin of the fast and thermal process in this sample.

Figure 4.14. Hysteresis response is measured at different modulating frequency. At high modulation frequency, the response is almost linear indicating this is a thermal process.

4.2.3 Tuning Cavity Periods

Since the FP cavity performance is dependent on parameters as the cavity reflectivities and effective absorption, the periods of the cavities are changed to understand the effects on the bistable properties. There samples were made with 4, 5 and 6 layers Bragg mirrors at the same porosity. Figure 4.15 (A-C) shows the Lorentzian fits of the samples transmission at different periods. The cavity resonane linewidths are observed to be 6.63 nm centered at 561.2 nm for the 4 periods, 5.6 nm peaks at 574.8 nm for 5 periods, and 4.39 nm at 551.8 nm with 6 periods which corresponds to Q-factors of 84.6, 102.6 and 125.7 respectively. These results indicate a clear improvement in the cavity performance expressing through shaper cavity resonance.
Consequently, this gives rise to a sharp increase in the intracavity intensity due to the fact that a high Q-factor is linked to high reflectivity cavity mirrors. Figure 4.15 (A-C) shows the transmission and reflection spectrum of cavity with 4, 5 and 6 layers mirrors. The spectra show a sharp increase in the absorption loss, i.e. reduction in transmission/reflections which indicate some sorts of trade off between sharper resonance modes and absorption from device physicality.

Figure 4.15. Lorentzian fits of the resonance transmission of cavities with (A) 4, (B) 5, (C) 6 periods Bragg reflectors
Figure 4.16. Reflection/Transmission spectra of samples with (A) 4 periods, (B) 5 periods and (C) 6 periods.

As a result, the onset bistable intensities observed in Figure 4.17 is shown to decrease with increasing numbers of layers. However, a trade-off is made between low switching intensities and damaged threshold. Since as the cavity periods increase, this will increase the cavity reflectivity mirrors as well as the sample thickness, now the samples become thicker which leads to a rise in the effective absorption constant. Hence, the result of rising absorption will cause the lowering of the damaged thresholds, and make samples more vulnerable to high excitation source. The onset intensities are shown to be at 1053 kW/cm$^2$, 884 kW/cm$^2$ and 231
kW/cm$^2$, while the damaged threshold is at 1676 kW/cm$^2$, 1268 kW/cm$^2$ and 330 kW/cm$^2$ for samples with periods of 4, 5 and 6 respectively.

**Figure 4.17.** Optical hysteresis of samples with (top left) 4 periods mirrors, (top right) 5 periods mirrors and (bottom) 6 periods mirrors.
V. DISCUSSIONS

1. Relationship between cavity porosities and the onset intensities and damaged thresholds

In order to observe the bistable effect, the minimum intensity that activates nonlinear transmission, i.e. switching intensity, needs to be smaller than the damaging threshold for the particular sample. This indicates that if the damaged threshold is lower than the onset intensity, the sample will become damaged before the bistability process happens as happened when using the 47% porosity samples. In the case of the samples with 60%, 71%, 80%, 90% and passivated samples with the same porosities, the switching intensity is much lower than the damaging intensity, so a large hysteresis is shown when examined those samples. The switching intensity seems to decrease as the porosity increases, while the damaging intensity increases with higher porosity sample.

![Graph showing onsets and damaged thresholds as functions of porosities](image)

**Figure 5.1.** Onset and damaged thresholds as functions of porosities

Our experimental data shows that the microcavity structure seems to improve the damaging threshold of porous silicon and lower the switching intensity when they are compared
to single layer structure [28]. Moreover, the trend of the threshold intensity with increasing porosity is different from previous work done on single layer at shorter wavelength. For single layer structure, it has been reported that the threshold intensity actually decreases with increasing porosities at 337 nm [192] which is due to an increase in the PSi absorption coefficient.

2. Characterizing the optical parameters of the cavity

A Fabry Perot cavity is defined by certain optical parameters such as the effective absorption loss $\alpha d$, the cavity reflectivities $R$ and the cavity thickness $d$. Thus, to understand its optical bistable property, we need to know these parameters which can be done by fitting equation (2.51) to the cavity transmission resonance, with $R$ and $\alpha d$ acting as fitting parameters. In this case, the cavity thickness is designed to be half of wavelength in length. The results are shown in Table 3.

\[
\tau = \frac{(1-R)^2 e^{-\alpha d}}{(1-Re^{-\alpha d})^2} \frac{1}{1 + F_n \sin^2 \left(\frac{2\pi nd}{\lambda}\right)}
\]  

(2.51)

<table>
<thead>
<tr>
<th>Porosities (%)</th>
<th>R (%)</th>
<th>$\alpha d$</th>
<th>nd</th>
</tr>
</thead>
<tbody>
<tr>
<td>60%</td>
<td>98.4</td>
<td>0.024</td>
<td>563.7</td>
</tr>
<tr>
<td>71%</td>
<td>98.8</td>
<td>0.02</td>
<td>567.5</td>
</tr>
<tr>
<td>80%</td>
<td>98.8</td>
<td>0.017</td>
<td>563.9</td>
</tr>
<tr>
<td>90%</td>
<td>98.8</td>
<td>0.0165</td>
<td>575</td>
</tr>
<tr>
<td>71% (passivated)</td>
<td>98.7</td>
<td>0.022</td>
<td>574.4</td>
</tr>
<tr>
<td>80% (passivated)</td>
<td>98.5</td>
<td>0.02</td>
<td>592.5</td>
</tr>
<tr>
<td>90% (passivated)</td>
<td>98.7</td>
<td>0.0195</td>
<td>584.3</td>
</tr>
</tbody>
</table>
The reflectivities of the mirrors using equation (2.51) for the 4, 5, 6 periods samples are deduced to be 97.6%, 98.8% and 99.5% which indicates an obvious increase in the cavity mirrors reflectivities. This is also confirmed by the fact that the cavity resonance linewidth becomes thinner as the samples periods increase. The absorption loss is also shown to increase with more periods. Overall, these results fit well with the transmission spectrum shown in Figure 5.2. To understand the effect of optimization of the FP cavity, a simulation of the intracavity intensity is conducted and is shown as following. From the plot, it is easily observed that even though with just a percentage increase in the cavity mirrors, the intensity inside the cavity shows an enhancement of almost 4 times. Such a high intensity can have a contradictory effect on the sample. On the one hand, it lowers the onset intensity since in order to exhibit bistability the device only requires a small excited intensity. On the other hand, such a high intensity can easily damage the sample especially in a high absorption background. These two effects are both observed in Figure 4.17. The samples tested show a lowering of onset intensities with increasing numbers of mirrors periods. However, they also show that the samples with the highest number of periods are much easily damaged than the other samples.

![Figure 5.2. Intracavity intensity with different periods.](image-url)
3. Simulation of optical hysteresis

In order to understand further the bistable effect, we modify the model used by Takahashi et al. [29] to extract the induced refractive index changes due to high power input pulse laser. First, we consider a nonlinear transmission process for a Fabry-Perot cavity as described by in equations (2.48)-(2.50)

\[
X = \left( \frac{2\pi dy}{\lambda} \right)^3 \frac{(1-R)(1+Re^{-ad})(1-e^{-ad})}{ad(1-Re^{-ad})^2} I_I
\]  
(2.48)

\[
Y = \left( \frac{2\pi dy}{\lambda} \right)^3 \frac{(1+Re^{-ad})(1-e^{-ad})}{ad(1-Re^{-ad})} I_T
\]  
(2.49)

\[
X = Y \left[ 1 + F_\alpha \sin^2 \left( Y_3^2 + \delta_o \right) \right]
\]  
(2.50)

where \( I_I \) and \( I_T \) are the input and output intensities respectively, \( R \) is the reflectivities of the front and back mirrors, \( \alpha \) is the absorption coefficient, \( \lambda \) is the laser wavelength, \( d \) is the cavity thickness and \( \gamma \) is the nonlinear refractive index coefficient, and \( F_\alpha = \frac{4Re^{-ad}}{(1-Re^{-ad})^2} \).

The nonlinear refractive index coefficient \( \gamma \) is characterized as shown in equation (2.52)

\[
\gamma = \beta \int_0^\infty I_{out}(t')e^{-\mu t'} dt'
\]  
(2.52)

where \( \beta \) is the nonlinear coefficient and \( \mu \) are chosen so that \( I/\mu \) is the transient carrier lifetime \( \tau_m \). \( \beta \) and \( \mu \) are the two fitting parameters.

Figure 5.3 shows the simulation model of the 71% porosity sample using information obtained from Table 3. The plot shows that the simulation agrees quite well with the experimental data. By applying the model to other porosities and the passivated sample, we obtain the values of \( \alpha \) and \( \mu \) which are shown in Table 4. The fitting data shows that the transient carrier lifetime \( \tau_m \) increases with increasing porosities, while the nonlinear coefficient \( \beta \) is almost
not dependent on the porosities. This can be thought as if the material is more porous, the nonlinear process is more persistent and corresponds to larger area of the hysteresis. Between 71% and 90% porosities, the nonlinear coefficient curves become a plateau while the lifetime increases dramatically. For the passivated samples, the value of $\tau_m$ is smaller than the value obtained from the unpassivated samples with the same porosities.

Figure 5.3. Simulation model of optical hysteresis.

Table IV. Fitting parameters for unpassivated and passivated samples at different porosities

<table>
<thead>
<tr>
<th>Porosities (%)</th>
<th>$\beta$ (ns)</th>
<th>$\mu$ (x 10$^6$)</th>
<th>$\tau_m$ (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>60%</td>
<td>6±4</td>
<td>(8±0.2) x 10$^6$</td>
<td>125</td>
</tr>
<tr>
<td>71%</td>
<td>8±4</td>
<td>(6.7±0.3) x 10$^6$</td>
<td>149</td>
</tr>
<tr>
<td>80%</td>
<td>9±4</td>
<td>(6.3±0.2) x 10$^6$</td>
<td>159</td>
</tr>
<tr>
<td>90%</td>
<td>10±4</td>
<td>(6.0±0.2) x 10$^6$</td>
<td>166</td>
</tr>
<tr>
<td>71%</td>
<td>8±4</td>
<td>(10.4±0.4) x 10$^6$</td>
<td>96.2</td>
</tr>
<tr>
<td>80%</td>
<td>8.5±4</td>
<td>(9.8±0.4) x 10$^6$</td>
<td>102</td>
</tr>
<tr>
<td>90%</td>
<td>14±4</td>
<td>(7.8±0.4) x 10$^6$</td>
<td>128</td>
</tr>
</tbody>
</table>

The transient lifetime $\tau_m$ confirms that the hysteresis observed is due to the transient effect, since the lifetime is longer than the excited pulse duration. The cavity response time $\tau_c$ is
in the order 10 fs which is also much smaller than both of the transient lifetime and the pulse duration. These three parameters $\tau_m$, $\tau_p$ and $\tau_c$ belongs to the moderately transient regime where $\tau_p \gg \tau_m \gg \tau_c$, and extreme transient where $\tau_m \gg \tau_p \gg \tau_c$

**Figure 5.4.** (Left) Nonlinear coefficient $\beta$ as a function of porosity, and (Right) Transient lifetime $1/\tau$ as function of porosity.

Another factor can contribute to the change in the nonlinear refractive index is the thermal effect. Weiss et al has showed that by heating up porous silicon multilayer structure to temperature as high as 373 K, the resonant mode of microcavity can exhibit a redshift of 3 nm [193]. However, the response time for such an effect is larger than 10 ms [194], while the samples were excited with laser of 150 ns pulse, and the transient lifetime obtained from the model was between 58.8 ns to 500 ns. Hence we would expect that if the thermal process was the dominant effect, the nonlinear response time would be in the order of milliseconds rather than nanoseconds. Moreover, such an effect would not be observable under nanosecond excited pulses.

In considering the key experimental findings of both the strong dependence of optical bistability on the porosity of the central cavity layer and on the surface passivation we suggest
that the observed optical nonlinearity is strongly linked to the surface states present in the internal surface of the porous silicon matrix. Previous studies have shown that the concentration of dangling bonds increases with increasing porosity [195]. The large concentration of dangling bonds with increasing porosity means that they can contribute significantly to the increase of the refractive index, which also happens in amorphous silicon [196]. This explanation also fits well with the experimental data of the passivated samples. The passivated PSi microcavities show significant increase in both the switching and damaged intensities. This is because the hydrogen bond now has been replaced by the carbon bond which has different properties, so they contribute differently to the nonlinear process.

4. **Pulse shaping simulation**

To study the dynamics of the temporal response of the input pulse during the bistability process, a series of pulses with different peak intensities and linewidths are simulated as the excited source using different transient lifetimes. With the 60% porosity sample, the simulation shows that if the input pulse becomes shorter, the hysteresis curves also become smaller. Figure 5.5 plots the hysteresis of the 60% porosity sample with different pulses input. The peak intensity is kept at the onset value which is equal to 846 kW/cm$^2$. The normal response pulse used in the experiment is 125 ns. As seen in the graph, when the pulse becomes 10 times shorter, the area of the hysteresis at this peak intensity also decreases, and eventually the response becomes linear at the picoseconds time scale. In order to observe some sort of hysteresis with picoseconds excited pulse, the peak intensity needs to be increased to 25 GW/cm$^2$. However this is impossible since the damaged threshold for this sample is at 1038 kW/cm$^2$. Thus, it can be concluded that the transient response is only limited in to nanoseconds. Moreover, if the pulse
becomes 10 times longer, the hysteresis is no longer observed, this also confirms the argument that this is an electronic response.

**Figure 5.5.** Optical hysteresis of the 60% unpassivated samples with different excited pulse width

Similarly, the 71% and 80% porosity samples also show decreasing hysteresis at fast modulation. If the input pulse is shortened to picoseconds scale, the 71% sample is expected to observe hysteresis at 17 GW/cm$^2$, and for the case of 80% the simulation shows that it can exhibit bistability at 14 GW/cm$^2$. Both of these values are larger than the thresholds, so the samples will be damaged before the intensities reach their peak values. However, it is interesting to note that the expected intensity for the picoseconds pulse decreases with increasing porosity of the active layer. Thus, the sample which is most likely to observe hysteresis with such short excited pulses is the 90% porosity, which is shown in the simulated pulse deformation plotted in Figure 5.6. The 90% sample also behaves in the same manners when excited by shorter pulses, i.e. the hysteresis becomes smaller.

To observe bistability when excited by picoseconds pulses, the 90% porosity sample requires a peak intensity of 13 GW/cm$^2$ which is below the damaged threshold. This value is smaller than intensity used by Maly *et al.* [180], which is about 2500 MW/cm$^2$, when they
observed hysteresis in PSi with picoseconds pulses. The difference can be explained through the fact that the sample studied is a FP cavity which has the ability to enhance the input intensity, while the sample studied by Maly et al were single layer structure.

Figure 5.6. Simulated pulse deformation of 90% sample with picoseconds pulse width

For the cases of the passivated samples, the 71% sample shows increasing bistability when the excited pulse is shorten by 10 times. However, the hysteresis is washed away when the pulse is reduced to the picoseconds time scale and it is replaced by the linear response. The simulation also shows that if the peak intensity reaches 17 GW/cm², the sample will be able to act as a bistable device. This required intensity is smaller than the observed thresholds meaning that the surface passivation process can help to speed up the switching process happening in the PSi device. The other passivated samples such as the 80% and 90%, however, act in a more similar behavior as those unpassivated with the same intensity. Through the simulation, the hysteresis obtained from the onset peak intensities become smaller as the pulse width becomes smaller, and the process becomes linear in the picoseconds time scale. However, the required peak intensities to observe hysteresis with picoseconds pulses seem to increase as the samples become more porous. For the 80% passsivated sample, the expected peak intensity is simulated to be 25 GW/cm², while in the case of 90%, the peak intensity is almost 45 GW/cm². This trend
indicates that the sample that is most likely to have picosecond switching time is the 71% passivated sample. Moreover, it also shows that surface passivation also links to the temporal response of the PSi FP cavity.

5. **Nonlinear refractive index**

Recall equation (2.52) describing the nonlinear refractive index affecting the FP cavity

\[ \gamma(t) = \beta \int_0^t I_{out}(t')e^{-\mu t'}dt' \]

Since the output pulse shape throughout the experiment is similar to a Gaussian function, then \( I_{out}(t) \) can be shown as

\[ I_{out}(t) = I_o + Ae^{\frac{(t-t_o)^2}{2w^2}} \]  

(5.1)

where \( A \) represents the maximum amplitude of the Gaussian function, \( t_o \) is the offset from the zero origin and \( w \) is the FWHM of the function.

Substitute \( I_{out}(t) \) we have the following

\[ \gamma(t) = \beta \int_0^t \left[ I_o + Ae^{\frac{(t'-t_o)^2}{2w^2}} \right] e^{-\mu t'}dt' \]  

(5.2)

If \( I_o \) is equal to zero, then

\[ \gamma(t) = \beta \int_0^t Ae^{\frac{(t'-t_o)^2}{2w^2}}e^{-\mu t'}dt' \]  

(5.3)

By integrating equation (5.3), \( \gamma(t) \) is equal to

\[ \gamma(t) = \beta Awe^{-t_o\mu} + \frac{w^2\mu^2}{2} \sqrt{\frac{\pi}{2\mu}} \text{Erf}\left[ -\frac{t-t_o+w^2\mu}{\sqrt{2w}} \right] \]  

(5.4)
where \( \text{Erf} \left[ \frac{t-t_0+w^2\mu}{\sqrt{2w}} \right] \) is the error function and is usually defined as

\[
\text{Erf}[x] = \frac{2}{\sqrt{\pi}} \int_{0}^{x} e^{-x^2} \, dx
\]  

(5.5)

The error function can also be approximated as following

\[
\text{Erf}[x] \approx \sqrt{1 - e^{-x^2/\pi(1+ax^2)}}
\]  

(5.6)

with \( a \) is equal to 0.14

Then \( \gamma(t) \) now can be approximated as following

\[
\gamma(t) = \beta A \text{w} e^{-t_0 \mu + \frac{w^2 \mu^2}{2}} \sqrt{\pi \frac{2}{e}} \left( 1 - e^{-\left( \frac{t-t_0+w^2\mu}{\sqrt{2w}} \right)^2} \right)
\]

(5.7)

A plot of the approximated form of \( \gamma(t) \) is given below with the onset peak intensity function of the 60\% porosity unpassivated sample. The actual function of \( \gamma(t) \) obtained by using numerical integration is also plotted in the same graph for the purpose of comparison. In this case, \( \beta = 6, A = 702.14, w = 1.25 \times 10^{-7}, t_0 = 2.96 \times 10^{-7}, \mu = 8 \times 10^6 \)

According to Figure 5.7, between 0 and 200 ns, the approximation curve indicates that the nonlinear refractive index decreases from an initial value to zero, and then increases after 200 ns. This means that the nonlinear process already happens even before the sample is excited, which does not fit with the physical observation. Comparing with the numerical integration method, the approximation curve fits better after 200 ns, which corresponds to \( t = \frac{t_0-w^2\mu}{\sqrt{2w}} \). Even though equation (5.7) does not represent the dynamics of the nonlinear refractive index
accurately, it can be useful to understand the dynamics between the excited pulse width $w$ and $\gamma(t)$. So as the pulses become faster, $w$ becomes smaller, which means that now the changes in $\gamma(t)$ happen at a much faster rate. As a result, if the time scale used to probe the sample is a few orders larger than the response of $\gamma(t)$, then only the horizontal part of the approximation curve can be observed, i.e. the refractive index has now become linear.

**Figure 5.7.** Nonlinear refractive index $\gamma(t)$ obtained through using numerical integration and the approximation function

As the peak values of excited intensity increases, the nonlinear refractive index also increases as shown in Figure 5.8. The refractive index initially linearly increases from zero, and as certain critical time, the value of $\gamma(t)$ increases dramatically till it reaches maximum value and starts to behave linearly again. Figure 5.9 studies the behavior of the input pulse as it passes through the cavity and the dynamics of the refractive index by plotting them on the same time scale. The time interval when the nonlinear refractive index exhibits sudden increase in its value overlaps with the rising of the input pulse. However, as the pulse starts to decay, $\gamma(t)$ continues to rise to its maximum value and then begins to increase linearly. Moreover, it is worth noting $\gamma(t)$ only behaves linearly when pulse deformation does not occur, i.e. if the refractive index
changes dramatically, it can induces from this that there is a nonlinear process happening in the cavity which causes the pulse deformation.

![Nonlinear refractive index at different peak intensities](image)

**Figure 5.8.** Nonlinear refractive index at different peak intensities

To understand the changes of the refractive as the pulse passes through the cavity, $\gamma(t)$ is plotted as a function of input intensity in Figure 5.10. The graph shows that the refractive index exhibits strong nonlinear properties by switching between the bottom branch and the top branch when pulse increases and decreases. The switching happens at when the intensity reaches its peak value, and when there is no pulse deformation, the curve is almost constant indicating a linear response. As the peak intensity is increased, the curve opens up more and more corresponding to large nonlinearity and so larger hysteresis.

![Dynamics of the refractive index and its relationship to pulse deformation](image)

**Figure 5.9.** Dynamics of the refractive index and its relationship to pulse deformation.
Figure 5.10. Nonlinear refractive index as function of intensities with different peak values.

Moreover, according to the fitting parameters, the highest nonlinear refractive index calculated is $6.67 \times 10^{-8}$ cm$^2$/W. Thus, the real part of the third order nonlinear susceptibility can be calculated based on equation (2.4)

$$Re \left( \chi^{(3)} \right) = 2n^2 \varepsilon_0 c \lambda \gamma$$

The value of $Re \chi^3$ calculated is in the order of $10^{-10}$ esu, which agrees with previous work. Also the value of $Re \chi^3$ of passivated samples is calculated to be in the order of $10^{-9}$ esu, which is similar to the results done in silicon nanocrystalline [197]. The increase in third order nonlinear susceptibility in the passivated samples is due to the fact that the linear refractive index $n$ now has increased because of the resonance shift induced by surface passivation as mentioned above.
VI. CONCLUSIONS AND FURTHER WORKS

In conclusion, this study has presented a detailed analysis of the optical bistability of mesoporous silicon microcavities with different porosities. The model shows that that bistable effect is due to a transient change in the nonlinear refractive index, and the process is predominately electronic. The threshold intensity has been shown to be increasing with increasing porosity, while the switching intensity is smaller with rising porosity. The transient lifetime seems to increase with increasing porosity. The passivated sample also behaves differently from the non-passivated sample, which indicates that surface area plays an important role in changing the nonlinear refractive index of mesoporous silicon. The role of the cavity structure has also been investigated and observed an enhancement in the intracavity intensity, and as a result, this lowers the switching intensity for cavity with higher numbers of layers. Other form of cavity optimization such as doping the sample with quantum dot also studied and shows that the sample exhibits both thermal and fast switching properties. However, the origin for such properties is still inconclusive.

A key challenge in this experiment that requires further improvement is tuning the shape of the bistable hysteresis so that it exhibits sharp switching rather than transient process as shown in the experiments, and so it is possible to use it as a more convenient form of bio-sensor without the need of a spectrometer. By observing the switching intensities through the oscilloscope as shown in the case of a normal optical bistability process, we can then easily detect any foreign objects existed inside the cavity. A possible solution for this is to create an asymmetric FP cavity, i.e. a cavity with less reflective front mirror and highly reflective back mirror. This form of cavity can be used to study the optical bistability process where the refractive index is altered due to both of the nonlinear refractive index and saturable absorption. Another possible solution
is to increase the length of the cavity due to the fact that the transient process is related to the cavity roundtrip time. Instead of making the cavity longer by adding more periods, which in turns would result in increasing the absorption loss, this can be done by making separate front and back mirrors with a mesoporous silicon etalon. However, there is a trade-off which is the large size will limit its application in nano-scale.

Further studies can also be conducted to have better understanding into the physical origin of the transient process. In this study, it is concluded that surface states play a large role in the bistable process; however an exact mechanism is still unknown. Moreover, the nonlinear refractive index under high intensity and short pulses in mesoporous silicon needs to be understood better. This can possibly be done by conducting an ellipsometric measurement which can provide more information about the nature of the nonlinear refractive index. Other important parameter such as the excited pulse width also should be studied in a more extensive manner such as conducting experiment with picoseconds pulses. This task was not accomplished due to the limited availability of such a laser. However, the project has proven to be a successful attempt into studying the order nonlinear process in mesoporous silicon microcavities which yield many interesting insights into the transient bistability process.
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