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REUSABLE PHOTONIC ENCRYPTOR ENABLED BY METAL-INSULATOR-METAL LAYERS

Bachelor's thesis
Faculty of Engineering and Natural Sciences
May 2021

ABSTRACT

Jesse Pietilä: Reusable photonic encryptor enabled by metal-insulator-metal layers
Bachelor's thesis
University of Tampere
Science and engineering
May 2021

A metal-insulator-metal (MIM) is a scalable optical structure. The cavity formed by planar continuous layers is of great interest as it can give controlled light transmission and reflection at the surface of the MIM structure. On the other hand, the generation of specific colors from the MIM structure is quite difficult, as the optical cavity can absorb a very narrow band and reflects the rest of the light. Nevertheless, in our study, we showed by precisely designing the MIM cavity we can achieve a peak in reflection instead of a dip. The peak in reflection means, the designed cavity is capable of reflecting a narrow band and absorbs the rest of the spectrum.

In this thesis, we use a MIM structure that has been made out of silver and silicon dioxide. We also create a pattern for the silicon dioxide layer. This pattern can be made either visible or invisible, depending on if the upper metal layer is attached or not. Polydimethylsiloxane (PDMS) surface is used to attach and detach the upper metal layer. The patternless structure is studied via computer simulations and the same type of MIM structure with a pattern in silicon dioxide is made in a laboratory. In simulations, we use a software called Lumerical FTDT Solutions what makes it possible to create three-dimensional models of the structure.

The thesis is divided into three parts. In the theoretical section, we study how MIM structures work as optical cavities. Furthermore, we find out how changes in the structure affect the color reflected from the surface. After this, the phenomena are studied with computer simulations. In this part, we study the reflectance of the structure. In simulations, multiple thicknesses are used so that the effect of the different thicknesses can be seen. All the results are presented via graphs and they are later compared to experimental results. In the experimental part of the thesis, we build a MIM structure that resembles the one used in simulations. We also add a pattern to this structure. With the experimental results, we prove that the MIM structure can be used as a reusable optical encryptor.

Based on the simulated results, it can be seen that thickening the silicon dioxide layer causes a redshift. This means that the feature of the cavity shifts to a higher wavelength. This can also be seen as a change in the surface color. Modifying the upper metal layer causes also changes, mostly in the amount of light reflected. The experimental result shares the same outcome with simulated results. IR spectroscopy has been used to experimentally record reflectance from the MIM structure that has been fabricated in the cleanroom. We can also come to the conclusion that the MIM structure can be used in optical encryption because the pattern in the silicon dioxide can be made invisible by removing the upper metal layer.

Keywords: Metastructure, MIM, reusable encryption mechanism, optical cavity, polydimethylsiloxane (PDMS) layer.

TIIVISTELMÄ

Jesse Pietilä: Metalli-eriste-metallipintoihin perustuva uudelleenkäytettävä optinen salaaja
Kandidaatintyö
Tampereen yliopisto
Teknis-luonnontieteellinen
Toukokuu 2021

Metalli-eriste-metalli (MIM) on skaalattava optinen rakenne. Tasomaisten jatkuvien kerrosten muodostama optinen kaviteetti on kiinnostava konsepti, koska se mahdollistaa hallitun valonläpäisyn ja heijastuksen MIM-rakenteen pinnalla. Erilaisten värien tuottaminen MIM-pinnalle on kuitenkin haastavaa, koska kaviteetti absorboi hyvin kapeaa aluetta valosta ja heijastaa loput pois. Tutkimuksessamme osoitimme kuitenkin, että tarkasti suunnittelemalla MIM-rakenne on mahdollista saada aikaan heijastuspiikki absorption sijasta. Heijastuspiikki tarkoittaa, että suunniteltu kaviteetti pystyy heijastamaan kapean alueen valon spektristä ja absorboi loput.

Työssä käytetään hopeasta ja piidioksidista rakennettua MIM-pintaa, jonka piidioksidikerrokseen työstetään kuvio. Tästä kuviosta voidaan tehdä silmin erotettava tai näkymätön riippuen siitä, onko ylin metallikerros paikallaan vai ei. Ylemmän metallikerroksen lisäämisessä sekä irrottamisessa käytetään apuna polydimetyylisiloksaanipintaa, johon metallikerros on kiinnitetty. Pintaa tarkastellaan ilman kuviota tietokonesimulaatioiden avulla, ja vastaava kuviollinen MIM-pinta valmistetaan laboratoriossa. Simulaatioissa käytetään Lumerical FTDT Solutions -ohjelmaa, jolla on mahdollista rakentaa kolmiulotteinen malli rakenteesta.

Työ jakautuu kolmeen osaan. Kirjallisuusosassa tarkastellaan MIM-pintojen toimintaa optisina kaviteetteina. Lisäksi selvitetään, kuinka muutokset pinnan rakenteessa vaikuttavat pinnan väriin. Seuraavaksi ilmiöitä tarkastellaan tietokonesimulaatioiden avulla. Tässä osassa tutkitaan muutoksia taittuneen valon aallonpituuksissa. Simulaatioissa käytetään useita eri paksuuksia pintojen kerroksille, jotta saadaan esiin niiden vaikutus taittuneeseen valoon. Tuloksista muodostetaan kuvaajia, joita voidaan verrata kokeellisiin tuloksiin. Työn kokeellisessa osassa rakennetaan simuloitua MIM-pintaa vastaava rakenne laboratoriossa, ja tähän pintaan lisätään pieni kuvio. Kokeellisten tulosten avulla todetaan, että pinta saadaan toimimaan uudelleenkäytettävänä salausmekanismina.

Simulaatioista saatujen tulosten perusteella huomataan, että piidioksidikerroksen paksuntaminen aiheuttaa punasiirron eli aallonpituuden kasvamisen. Tämä voidaan havaita pinnan värin muutoksena. Myös ylemmän metallikerroksen kasvattaminen aiheuttaa muutoksia, lähinnä taittuneen valon määrässä. Kokeellisten tulosten avulla päästään myös samaan päätelmään. Kokeelliset tulokset on mitattu laboratoriossa valmistetusta MIM-pinnasta. MIM-pinnan todetaan myös toimivan datan salauksessa, koska siihen rakennettu kuvio saadaan sekä näkyviin että piilotettua lisäämällä päällimmäinen metallikerros.

Avainsanat: Metapinta, MIM, uudelleenkäytettävä salausmekanismi, optinen kaviteetti, polydimetyylisiloksaanipinta

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1 INTRODUCTION

As technology keeps evolving, the need for more advanced security features becomes more significant. Many of the existing security mechanisms will be compromised as time passes and computer computing power increases. To prevent forgeries such as counterfeited currency or documents, it is important to invent new methods to authenticate valuable products. One way of achieving this is by using what is called optical security features. These techniques rely on visual changes in materials when interacting with a certain type of electromagnetic radiation. The aim of optical security features is to produce predetermined visual effects under specific stimuli. For example, putting a banknote under ultraviolet light reveals several different types of images and numeric codes. To make copying these types of pieces difficult, the optical features should be hard to mimic and reproduce. [1][2]

There are numerous ways to use optical phenomena to hide or encrypt data. Many of the existing security features utilize concepts like photonic crystals, dielectric metasurfaces, and holograms [1]. However, this bachelor's thesis mainly focuses only on a simplified planar structure consisting of metal-insulator-metal (MIM) cavities. Metamaterials are made from nano-sized surfaces, whose thicknesses are a lot smaller than the wavelengths of light. Because of this, they can be used to affect the behavior of electromagnetic waves. [3] These types of materials do not normally occur in nature and share promising results due to their unique optical properties such as tunable reflectance. Metal-insulator-metal metasurfaces exhibit unique properties that make them useful in security applications. For example, by using MIM surfaces it is possible to produce patterns large enough to be seen with the naked eye or a camera, and the visibility is not generated by using micro- or nano-scale shapes. [1] Creating these kinds of surfaces also requires a laboratory environment, specific materials, and a complicated process, which makes them an ideal security adaptation.

The different properties of the structure can be changed, to produce distinct colors using MIM structures. By affecting the thickness of the insulator layer, it is possible to change the reflectance of the material. Furthermore, by modifying the thickness of the top metal layer, it is possible to produce more distinguishable colors. [1]

The goal of this thesis is to study how changes in the structure affect the optical features of the MIM structure. The main focus is on creating visible color changes by modifying the layers' thicknesses. We also show that it is possible to create pre-determined patterns into the structure by making partial changes in the thicknesses of the layers. To study these effects, we use computer simulations to measure the reflectance of the MIM structure with different layer thicknesses. Later, it will be shown experimentally how to utilize the MIM structure to create a security application. Here a predetermined pattern is encrypted into the insulator layer. After this, a polydimethylsiloxane (PDMS) patch, coated with a thin silver layer, is added on top of the structure to decrypt the data. The experimental results are then compared with the simulations and all results are presented via graphs and pictures.

Chapter 2 is going to focus on the basics of MIM structures and cover the theory about the subject. In the 3rd chapter, we present simulated results by using computer software. The 4th chapter is going to explain how the physical sample is made step by step in a laboratory. In the 5th chapter. We present experimental results measured from fabricated samples and the 6th chapter summarizes all the results and thoughts.

2 THEORETICAL OVERVIEW OF MIM-STRUCTURES AS COLOR FILTERS

There are multiple ways to produce colors on surfaces, these mechanisms are divided mostly into two categories: pigmentary and structural coloring. As pigmentary coloring relies on pigments that are insoluble in water, structural coloring uses micro- and nanostructures that are small enough to affect the refraction of the visible light. [3] Structural coloring can be frequently seen amongst many animals in nature, for instance in the feathers of birds or butterflies' wings [4]. Structural colors that are caused by reactions between visible light and tiny metallic designs are called plasmonic colors. Plasmonic color filters are categorized by their structure. There are filters that rely on different types of hole, rod, or metal-insulator-metal designs. [3] This thesis focuses only on the latter, i.e. the MIMs.

2.1 The basics of MIM-structures

Metal-insulator-metal structures can be used as plasmonic color filters because they form a structure called an optical cavity. Optical cavities are structures that use optical components, usually mirrors, to trap a beam of light inside a closed path. In the case of MIM structures, the optical cavity is constructed from an optically thick bottom mirror and a thin top metallic layer. These two metallic layers are separated by another layer that is made out of a dielectric material. To be more precise, MIM structures form a Fabry-Pérot interferometer. [3] This is a type of optical cavity that is made out of two parallel surfaces that can filter out certain wavelengths of the light illuminating on their surfaces. [5, p. 39]

The basic principle of the MIM structure is that the incoming ray of light hits the thin top metallic film. When the light interacts with the MIM cavity and then reflects from the bottom mirror, it reflects a specific narrow band and absorbs the rest of the spectrum of incident light. Specifically, which narrow portion of wavelength it will reflect, depends on the design of the MIM cavity. In the theoretical case, the bottom mirror is considered to be optically thick enough to reflect all the light back. Also, the top mirror should be really thin so it can act as an absorber. By using Snell's law, it is known that when light travels from a less optically dense material into a more dense material, the light will refract towards the normal. Conversely, when light moves from a more optically dense material into a less dense material, the light refracts away from the normal. [6] Because of this, the MIM structure traps some of the light between the mirror layers and as a result filters out some wavelengths of the light. This effect is presented in Figure 2.1.

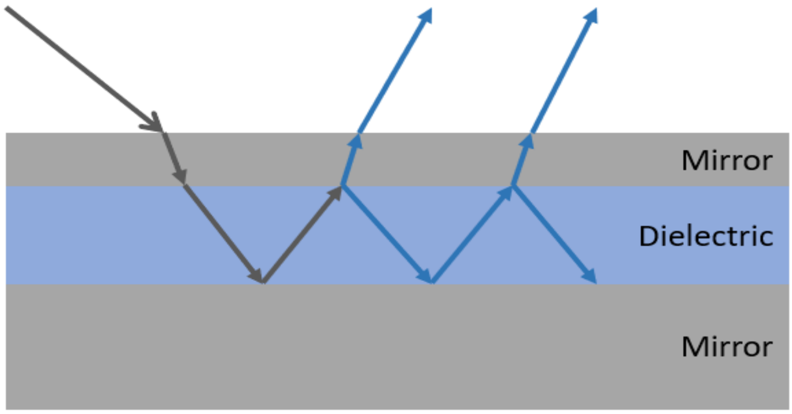

Figure 2.1. *A beam of light interacts with a MIM structure that consists of a thin top metallic layer, dielectric layer, and an optically thick bottom mirror.*

The optical response of these types of MIM structures depends on the materials and thicknesses of each layer. In other words, by changing the properties of the layers, it is possible to determine what wavelengths of light the material filters out. It is also possible to stack more layers on top of each other and filter out multiple wavelengths of light. By using several metal-insulator layers and tuning their thicknesses, structures that are near-perfect absorbers can be built. These types of materials can absorb most of the spectrum of visible light. [7] However, in the case of simple MIM structures, the changeable parameters are the material of the top and bottom metal layers, dielectric material, thickness of the dielectric, and the thicknesses of the two metal layers. In this thesis, silver (Ag) is used as the material for both top layer and bottom mirror. Because silver works as a great reflector for the visible spectrum of light; being able to reflect near 95% of it [8]. Silicon dioxide (SiO_2) is used for the dielectric material due to its lossless and transparent structure. This way the dielectric layer will reflect as little light as possible. It is also important for the bottom mirror to reflect as much light as possible, so it needs to be optically quite thick. Here a 100 nanometer (nm) silver layer is used for this purpose in the simulations, and also in the experimental part of the thesis. Now the only parameters left are the thicknesses of the upper silver surface and the dielectric material. By changing these, the color reflected from the structure can be adjusted. Changing the thickness of the dielectric redshifts the reflection spectrum, affecting the shade of the color created. And by increasing the top silver layer the colors can be made sharper. Up to this point, the layers have been considered to be evenly thick, so that there have been no variations in the thicknesses inside layers. However, if the thickness of the dielectric material is changed partly, it would be possible to create a pattern that would be visible to the naked eye. This is a result from the fact that optical cavities with different mirror distances absorb different wavelengths of light. Due to this, the optical cavities also produce different surface colors, making it possible to create color changes into the surface.

2.2 Reversible decryption of patterns by using MIM structures

It is possible to utilize the optical properties of MIM structures to create a decrypted pattern onto the MIM surface. Earlier it is realized that by modifying the thickness of the dielectric layer, the color of the surface can be changed. Now to demonstrate decryption of pattern onto the surface, few nanometer deep patterns are created in a dielectric layer using photo-lithography. Patterns are invisible to naked eyes. After this, a top metal layer is applied on top of the dielectric to reveal the hidden pattern. This way a visible pattern can be created onto the surface. The variating thicknesses of the dielectric layer will filter out different wavelengths of light and produce changes in the colors. However, the pattern will only be visible when the top mirror layer is applied. Without the top absorber, the light passes through the insulator layer and reflects only from the bottom mirror, making any changes in the insulator invisible. Basically, without the thin top metal layer, the reflection spectrum of the structure will be similar to the spectrum of the bottom mirror. These instances are presented in Figure 2.2.

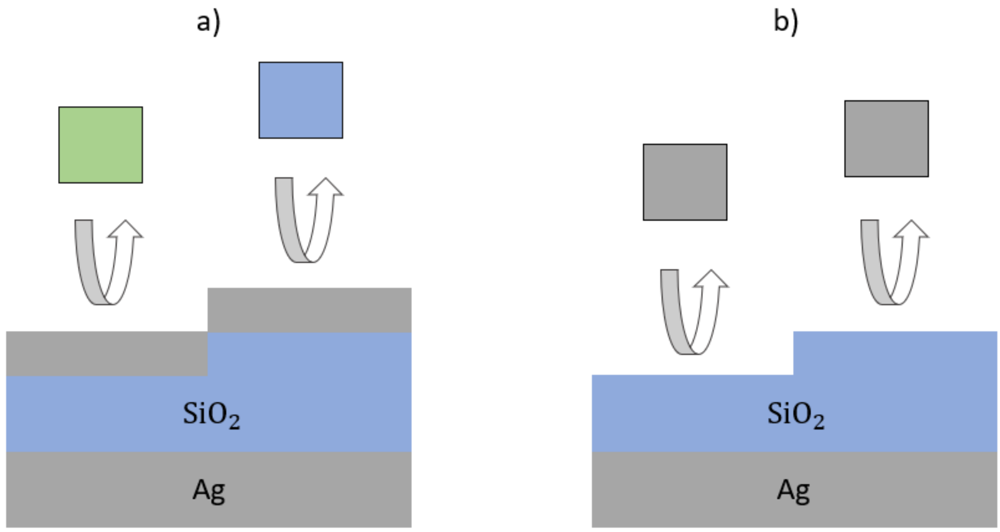

Figure 2.2. *Cross-section pictures for a) MIM structure consisting of an optically thick silver layer as the bottom mirror, variating SiO_2 layer as dielectric and a top silver layer as an absorber, and b) The same construct but without the upper silver layer. This figure is based on Figure 2. in [1].*

By utilizing these types of optical cavities, encrypted patterns can be generated onto the insulator layer of the structure. The patterns can be decrypted by applying the top mirror layer, making the pattern visible to the naked eye. The thin metal layer can be deposited directly on top of the insulator layer, but this turns out to be inconvenient, as the process is irreversible. To make reversible decryption possible, the absorber needs to be applied to a separate layer that is made out of transparent elastomeric material. In this thesis, we use polydimethylsiloxane as the elastomeric material. This way we can apply the elastomer patch with the top mirror layer onto the structure and also remove it without damaging the layers. Now the pattern can be encrypted and decrypted multiple times. Examples of the structure with and without the elastomer patch are presented in Figure 2.3.

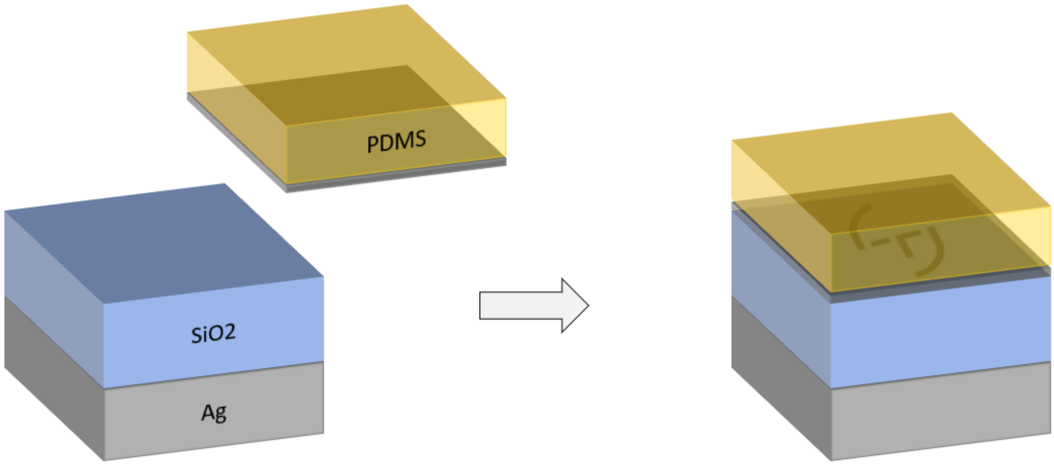

Figure 2.3. *Reversible decryption of a Tuni-logo pattern that is produced by making small changes to the* SiO_2 *thickness. The patter becomes visible after sticking on the elastomer patch coated with a silver layer. The figure is based on Figure 1. in [1].*

In realistic models, there is also a need for a protective layer between the thin metal layer and the insulator layer. Otherwise, the thin mirror layer might be peeled off and transferred onto the insulator layer, making the optical cavity permanent. [1] However, in later simulations and experimental parts, we are trying to reproduce a structure that mimics the one in Figure 2.3.

2.3 Adding periodic holes onto the MIM structure

By using a simple MIM structure and a PDMS patch, it is possible to create visible color changes and patterns to the surface. This satisfies our needs when studying MIM structure's usage as a security feature. However, when applying this theory and creating real-world applications, it would be important to enhance the optical output of the structure. In order to meet high application requirements, some qualities such as color brightness and saturation need to be improved. This can be achieved by making small modifications to the structure's surface.

From an application standpoint, it would be important to produce a wide range of colors with good brightness. By achieving high reflection for three primary colors, it would be possible to utilize the cyan-magenta-yellow system (CMY). By using the CMY system, it is possible to create thousands of colors that the human eye can recognize. One way of producing these colors with high saturation is to utilize periodic hole arrays.

By making small holes into the MIM structure, more precisely, into the top metal layer it is possible to affect the absorbed light. These holes are a lot smaller than wavelengths of visible light and they are in triangular-lattice formation. The hole structure will excite surface-plasmon polaritons (SPPs). SPPs are electromagnetic waves that move along metal-insulator surfaces. SPPs are in either infrared or visible light -frequency. In the MIM, the dielectric layer between the metal mirrors couples the SPPs on two metal-insulator interfaces. This creates a magnetic dipole resonance. With both electric and magnetic dipole resonances in the structure, the impedance of the free space will be matched. This effect results in near-perfect light absorption for the top metal layer. [9] Some studies also propose that the periodic hole structure results in a greater wavelength interval when the holes are in triangular formation rather than in square formation. This means that the triangular formation will reduce the color cross-talk and result in richer colors. [10][11]

The change in absorbed wavelengths can be achieved by modifying the size and periodicity of the holes. [9] In other words, it is possible to change the surface color and brightness by variating the hole size and periodicity. This type of color creation method can be used to improve the optical properties of MIM structures. It is also an optimal method for example color printing because with the nano-sized holes it is possible to print small patterns.

3 MIM STRUCTURE SIMULATIONS

In order to see how the MIM structure behaves under a certain type of stimuli, we are going to simulate the structure by using a software called Lumerical FDTD Solutions. With Lumerical we can build a 3d-model of the structure and simulate its interactions with a light source. The goal is to build a structure that resembles the one in Figure 2.3. Although, we do not simulate the small Tuni-logo, just the flat MIM structure with the PDMS layer. The results for the reflectance of the structure are later presented in graphs. In later chapters, we also compare the simulated results to experimental results. The experimental results are measured from the samples that are created in a laboratory.

We start simulations by making the lower Ag layer of thickness 100 nm. Lumerical uses different sources for the refractive index of materials and the options are Handbook of Optical Constants of Solids [12], CRC Handbook of Chemistry & Physics [13], and Optical Constants of Noble metals [14]. In this case, the CRC Handbook is chosen as the source for the silver. On top of the silver, we place the silicon dioxide layer which thickness variates. The four thicknesses used are 110 nm, 120 nm, 130 nm, and 155 nm. In the case of silicon dioxide, there is only one option for the material. This is the material whose refractive index is based on the Handbook of Optical Constants of Solids. After the SiO_2 is applied, the next step is to add the top silver. Here we use the same material choice as with the lower silver layer. The top silver layers thickness also variates. The thicknesses are 10 nm and 20 nm. After the MIM structure is ready, we add the PDMS layer. The thickness of the PDMS is set to 2000 nm. The material for the PDMS is a user-defined dielectric material, and we set the index value as 1.4.

When the MIM structure and PDMS are ready, the next step is to add a light source and an FDTD region. The source is set to plane wave and the FDTD region is delimited so that the region is inside the structure. Next, we determine the boundary conditions for the FDTD region. The x-axis is set to periodic as well as the y-axis. However, the z-axis is set to PML, which stands for a perfectly matched layer. This has a nearly perfect absorption and it also minimizes reflections. The settings for mesh are also determined. At this point, we also add a reflection monitor that measures the reflectance of the material. This is placed above the light source so that the monitor measures the reflected light.

Lastly, the wavelength interval is determined. The range for wavelength is set from 350 nm to 750 nm.

After the structure and all settings are ready, the simulation is started. The goal is to simulate all possible variants. This means that the simulation is run multiple times using different thicknesses for top silver and silicon dioxide. Simulating all the different variants gives us eight simulation runs. The simulation measures the reflectance of the material as a function of the wavelength. The results are presented in Figure 3.1.

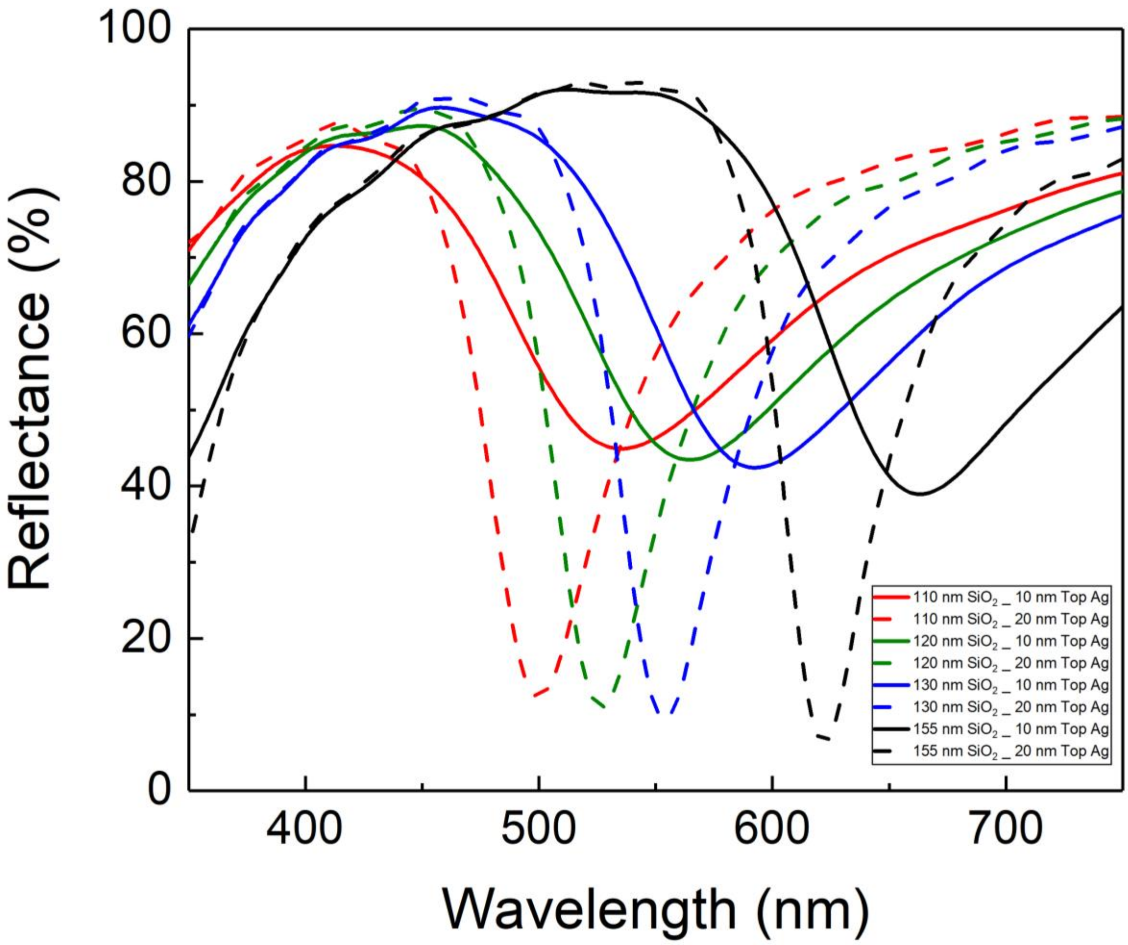

Figure 3.1. *Simulated reflectances for MIM structure as a function of wavelength. Different SiO_2 thicknesses are marked with different colors. Also, 10 nm top Ag layers are marked with solid lines and 20 nm layers are marked with dashed lines. All the simulated MIM structures have a lower Ag layer of 100 nm and a 2000 nm PDMS layer.*

Figure 3.1 shows the effect of different layer thicknesses on the reflectance of the structure. We can observe that thickening the SiO_2 layer indeed causes a redshift what can be seen as a change in the reflected light's color. The redshift happens with both thicknesses (10 and 20 nm) of the top Ag layer. This can be seen by comparing the dashed lines together, and also the solid lines together in Figure 3.1. By using thicker SiO_2 the lines move towards a bigger wavelength as this is what a redshift means. By thinning the SiO_2 layers we can cause an opposite effect where the wavelength decreases. This is called a blueshift.

If we compare the dashed lines to the solid ones in Figure 3.1, we can also see a redshift. This means that changing the upper Ag layer also has an effect on the wavelength and so by the color of the structure. This happens because the upper Ag layer also has an impact on how the optical cavity works. Thickening the top mirror affects what wavelengths of the light are passed through the surface and also what wavelengths escape or are reflected from the cavity.

By analyzing Figure 3.1, we also notice the difference in reflectance when moving from 10 nm top Ag to 20 nm top Ag. We can notice that with a thicker top layer, the surface absorbs more of the light, in fact over 60% more. This happens because, with a thicker top metal layer, the optical cavity traps more of the light inside itself. The thicker top mirror reflects more of the light away from the surface, resulting in less light passing through the top mirror. However, due to Snell's law, the light will hit the top mirror at a smaller angle when exiting the structure than when entering the structure. This leads to more of the light getting trapped inside the structure when using a thicker top mirror. This effect was presented in Figure 2.1.

The simulated results prove that by changing the thickness of the SiO_2 layer, we can achieve different colors for the surface. The next step is to replicate a similar structure in a laboratory and variate the thickness of the SiO_2 layer to produce a visible pattern.

4 CREATING THE MIM STRUCTURE AND AN ELASTOMER PATCH IN THE LABORATORY

Creating the MIM structure is a multi-step process that requires an environment containing a very low level of particles, such as dust. The samples created for this thesis are made in a cleanroom that is located on Tampere university’s campus. In order to test various thicknesses for the silicon dioxide layer, four main samples are created. Two of the samples are going to have a 120 nm SiO_2 layer and two are going to be with a 130 nm SiO_2 layer. All the samples have a bottom silver layer with a thickness of 100 nm. These samples will be later tested with different thicknesses of the top Ag layer, which is attached to the PDMS patch, to see what variation produces the best visual effects.

4.1 Silver deposition onto the glass surface

The first part of creating the silver-SiO_2-silver surface is to prepare the base where to deposit the lower silver coating. Here glass samples, roughly the size of one square centimeter, are used. These glass surfaces are first cleaned properly so that there are no pollutant particles that would harm the silver coating.

For the deposition process, we use an instrument called an electron beam evaporator, which consists of two main chambers, operated under a high vacuum. The glass samples are mounted to a sample holder and placed inside the upper chamber. After the chambers have reached their target pressures, a laser is used to shoot an electron beam to the target metal, located inside the lower chamber. In this case, the metal is silver. The electron beam forces atoms from the metal to change to gas form. After this the gas moves to the upper chamber and transforms back to a solid state, creating a thin layer of silver on top of the glass sample. The thickness of the silver layer can be predetermined using the machine’s settings. With our samples, the lower silver layer is 100 nm thick.

After taking the samples out of the evaporator, it is noticeable that their surface has changed from being transparent to being highly reflective. Now the samples work as a small mirror reflecting light very well. The next step in creating the MIM structure is to produce the silicon dioxide coating on top of the silver mirror.

4.2 A Silicon dioxide deposition onto the silver surface

After the silver deposition has been completed successfully, the next step is the SiO_2 deposition. Here the same type of electron beam evaporator is used to produce the silicon dioxide layer as it was used with the silver. The difference is that now, instead of using metal, we are going to evaporate SiO_2 which is a dielectric material. With this deposition, the temperature needs to be taken to account more precisely. This is why a temperature sensor is mounted to the sample holder so that the temperature can be monitored during the evaporation procedure. Before evaporating the actual samples, a couple of calibration runs are necessary, so that the thickness of the SiO_2 layer can be set as close as possible to the ideal number. During the calibration process, we evaporate random samples that already have a thin reflective coating on them and the SiO_2 is evaporated on top of that. The SiO_2 thickness should be as close as possible to the thickness we want on to the actual samples. After each calibration run, the SiO_2 layer is measured with an ellipsometer and the tooling factor is changed in the evaporator machine. When the values are in the desired range, we can continue with the real samples.

After the calibrations are completed, we move on to evaporating the actual samples. Because there are two sets of samples with different ideal SiO_2 thickness, two evaporation runs are needed. An extra sample with a thin reflective layer is also placed in the machine with the other samples so that the exact thickness of the SiO_2 layer can be measured with the ellipsometer. Now the evaporation process is going to be carried out the same way as the metal evaporation procedure, except we pay close attention to the temperature during the evaporation.

After the dielectric evaporations are completed, the extra samples are used to measure the SiO_2 thickness with the ellipsometer. We can also see that the extra samples have a bit different color than the silver-SiO_2 samples, due to different bottom layer thickness and material.

4.3 Creating the stencil for photolithography

To produce the predetermined pattern onto the dielectric layer, a photolithography method is used. This offers us a way to make high-resolution and large-scale designs, but this method is limited by the accuracy of the laser cutter, that we use to cut the stencils. In our case, it is important to choose a pattern that fits onto the one square centimeter-sized sample and is visible with the naked eye.

We ended up testing several different types of materials and patterns. After this, an opaque plastic sheet is chosen as the material and for the pattern, we decided to use two different ones: a logo of the Tampere University and the word meta. All of these stencils were produced in Tampere University's Fablab with a laser cutter.

4.4 Producing the polydimethylsiloxane patch coated with a silver layer

Creating the PDMS patch is a process of its own. This takes place in Tampere University's redlab-laboratory, where we start by weighing and mixing the PDMS monomer and hardener into a small bottle. PDMS-liquid is made out of monomer and hardener in a ratio of 10:1. After the liquid is properly mixed, the bottles are placed inside a vacuum. In this way, the unwanted air bubbles can be separated from the mixture.

As the PDMS-liquid is in a vacuum, we start producing the thin silver layers, that are going to be transferred onto the PDMS. Before depositing the silver, we are going to place the glass samples in a spin coater and put a couple of drops of polymethylmethacrylate (PMMA) liquid on top of them. PMMA, also known as acrylic, is a synthetic thermoplastic that is widely used, for example in aquariums due to its strong build and transparent nature [15]. The spin coater spreads the PMMA which will serve as a sacrificial layer. This is done for all samples. After this, we are going to deposit two different silver thicknesses onto the glass-PMMA samples, using the same e-beam metal evaporator that we used for the bottom mirror. The thicknesses of the top layer are 10 nm and 20 nm.
After the PDMS-liquid is ready to use and the silver layers have been prepared, the silver samples are placed in the chamber to deposit MPTMS monolayer on it. This improves adhesion between PDMS and Ag. In the end, on top of these samples, we spin-coated a thick PDMS polymer layer. After the spin coater, the samples are placed on a hot plate, where the PDMS cools down and solidifies. Now there is a structure that contains layers of glass, PMMA, silver, and PDMS. To get the PDMS layer with silver coating, the PMMA

needs to be dissolved, so that the silver and glass layers will separate. This part is done by using 1-methyl-2-pyrrolidinone. There is also an option to use other substances to dissolve the PMMA, but 1-methyl-2-pyrrolidinone is ideal in this case. This is because the thin silver layer will float in it, unlike for example in acetone. This helps the separation process, as the glass part will sink in the liquid and the PDMS-silver patch will end up floating.

The reason why the PMMA had to be used in order to create the PDMS-silver patch is that the PDMS needs to be placed on the silver layer. If this would be done the other way around, the PDMS layer would need to be put inside the metal evaporator. This is something the PDMS layer cannot handle due to the extreme conditions inside the machine. The silver layer also needs a base because it is very thin, but it cannot be deposited straight onto the glass layer. Otherwise, there would be no way to separate the glass and silver layers. This is why there is a soluble PMMA layer between the glass and silver.

5 EXPERIMENTAL RESULTS

5.1 Placing the PDMS patch onto the metal-insulator structure

After the MI structure with a pattern embossed in the insulator layer and the PDMS layer are ready, it is possible to test their functionality as an optical security method. Before placing the PDMS coated with silver, the silver-SiO_2 structure looks like a mirror. The structure reflects most of the light back and the pattern in the SiO_2 is not visible to the naked eye. The PDMS layer can be placed on top of the metal-insulator structure by using tweezers. After this, we can see if the pattern in the SiO_2 becomes visible.

Out of all the samples created, two variations seemed to produce the best outcomes. Having the 10 nm Ag layer with the PDMS on top of the 120 nm and 130 nm SiO_2 layers created the most visible patterns to the naked eye. The 20 nm Ag layer did not produce a very visible pattern. The reason could be that the layers might be in fact a bit different size than predicted.

Pictures of the two working samples are presented. The MIM structure with 120 nm SiO_2 layer and 10 nm Ag layer is presented in Figure 5.1. Furthermore, the structure with 130 nm SiO_2 layer and 10 nm Ag layer is presented in Figure 5.2. Having different SiO_2 thicknesses in two samples results in the colors also being different. This can be seen by comparing Figure 5.1 to Figure 5.2.

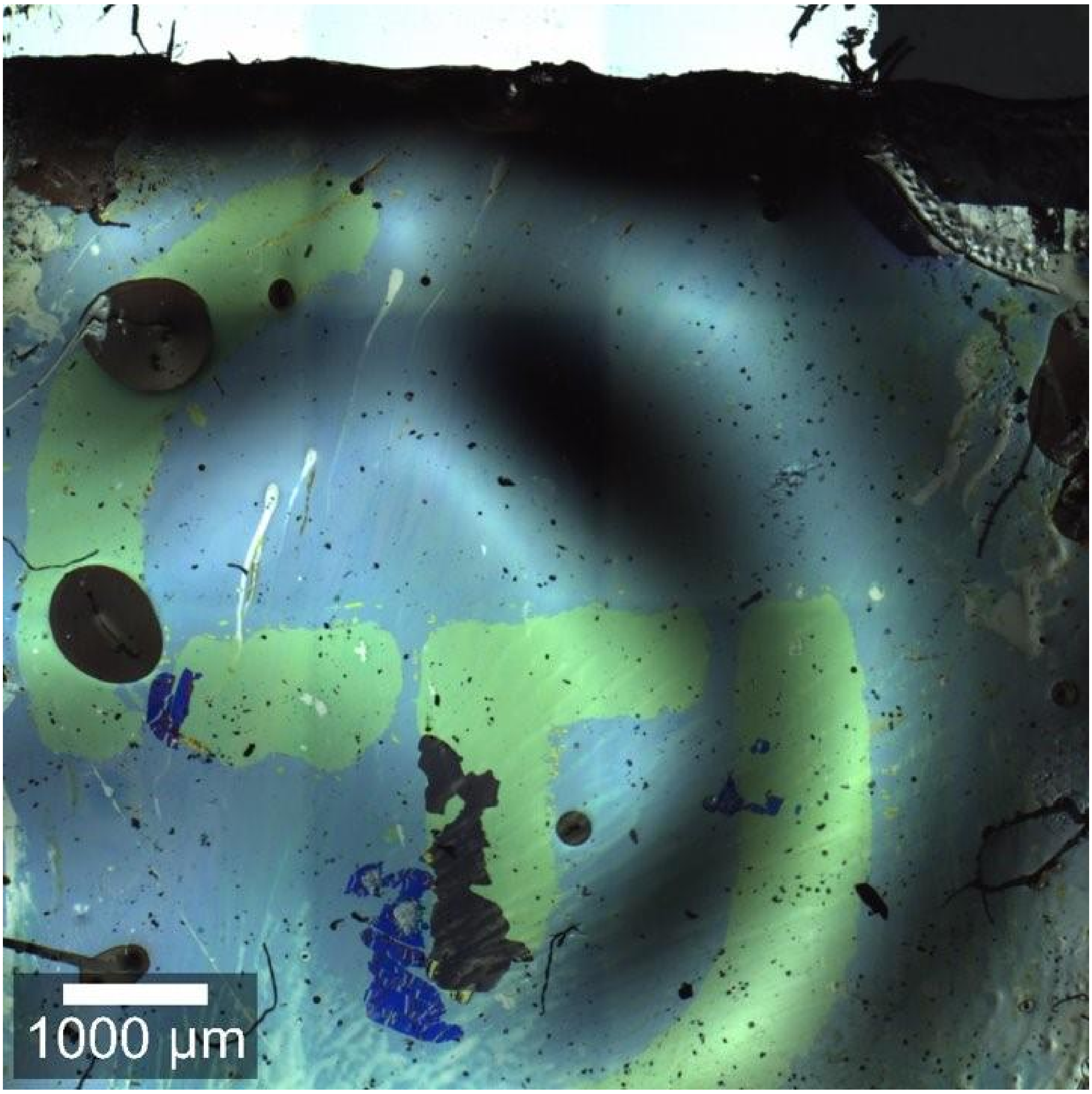

Figure 5.1. *Microscope picture of the MIM structure with the PDMS patch. Silicon dioxide thickness is 120 nm, and the top silver thickness is 10 nm. Picture taken by Dipa Ghindani.*

As seen in Figure 5.1, the green Tuni-logo-pattern is clearly visible to the naked eye and a camera. However, there are a noticeable amount of unwanted particles inside the structure. This is probably due to the PDMS layer being made outside the cleanroom, compromising the layer’s purity. There are also noticeable dark circles between the PDMS patch and the insulator layer. This tells that the PDMS layer has not stuck entirely into the insulator layer, leaving air bubbles between the two layers. The performance of the MIM structure could have been improved by adding a protective coating to the structure. The picture is taken through a microscope and the scale is presented in the lower-left corner.

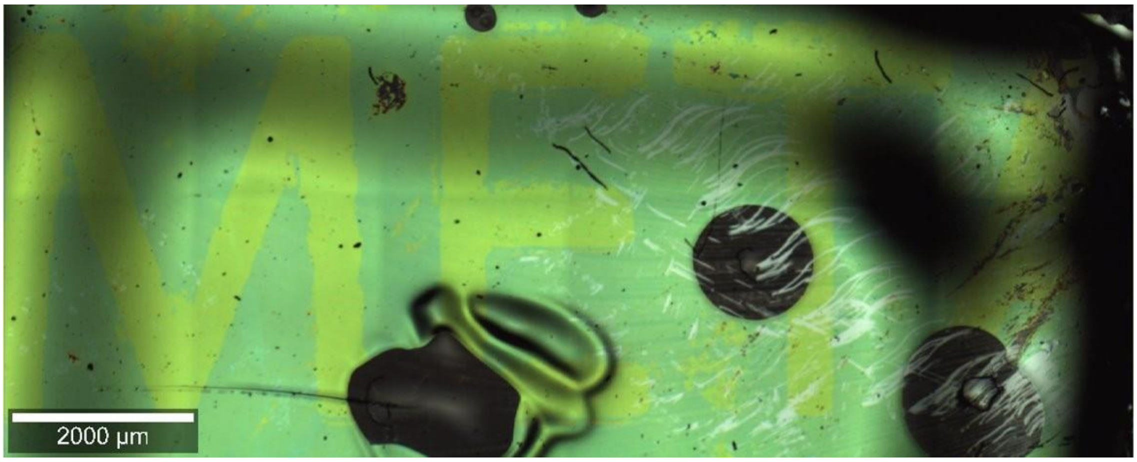

Figure 5.2. *Microscope picture of the MIM structure with the PDMS patch. Silicon dioxide thickness is 130 nm, and the top silver thickness is 10 nm. Picture taken by Dipa Ghindani.*

In Figure 5.2, we can see the word “META” being visible over a green background. There are also some air bubbles and a major hole in the structure. The hole is in the 10 nm silver layer and shows how sensitive the thin layer is to damage.

All in all, the patterns seem to be clearly visible after the PDMS has been stuck on. The PDMS can also be removed with tweezers, making the pattern invisible again. Furthermore, the PDMS patch can be used multiple times. This proves that the MIM structure can be used as a reusable optical security method.

5.2 The reflectance of the physical sample

After the physical sample has been tested, we measure the reflectance of the structure with different layer thicknesses. The goal is to get similar types of results as with the theoretical simulations. Here we use only the 120 nm and 130 nm thicknesses for the SiO_2 and 10 nm and 20 nm thicknesses for the top silver, as these are the samples we created in the laboratory. The results are presented in Figure 5.3.

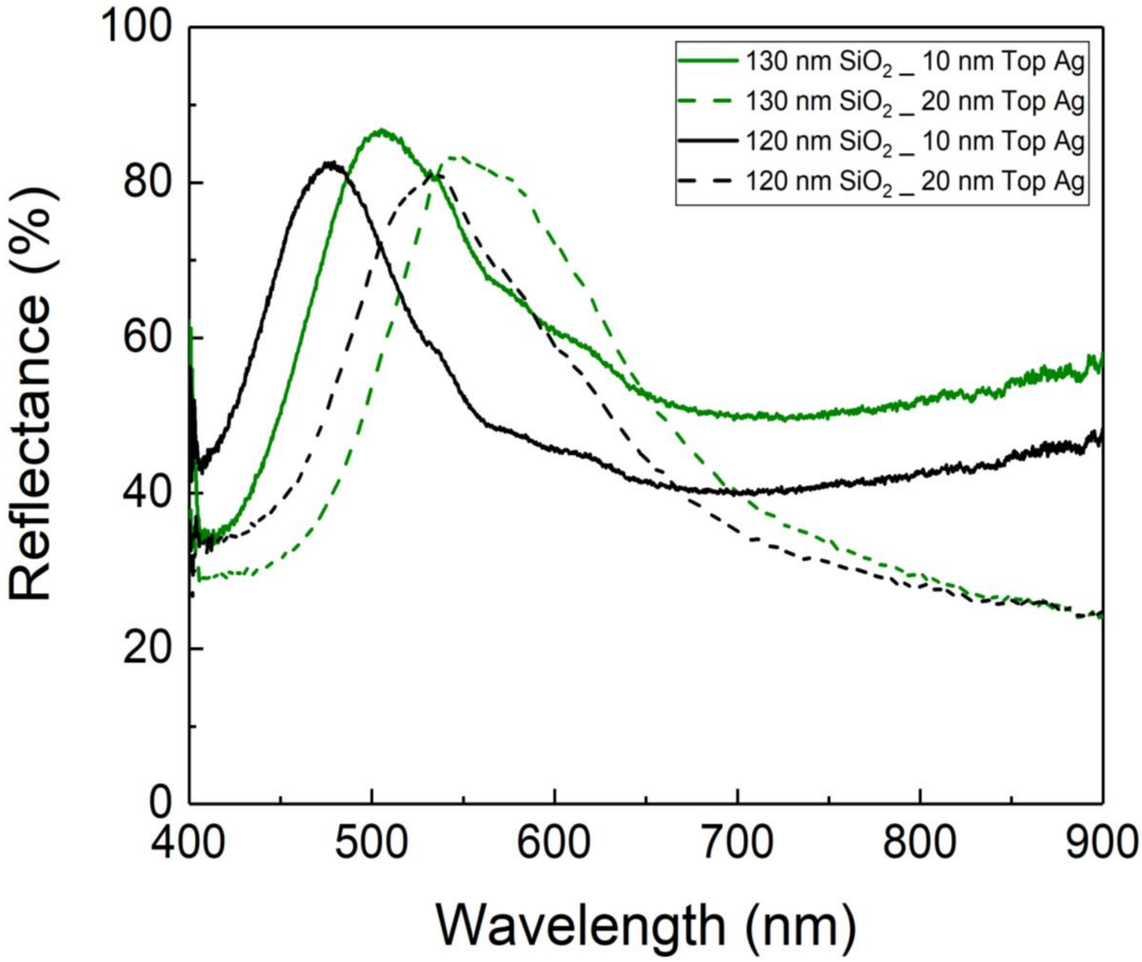

Figure 5.3. *The measured reflectances for the laboratory sample. Different SiO_2 thicknesses are marked with different colors. Also, 10 nm top Ag layers are marked with solid lines and 20 nm layers are marked with dashed lines. All the MIM structures have a lower Ag layer of 100 nm and a 2000 nm PDMS layer.*

Figure 5.3 shows the expected redshifts when increasing the layer thicknesses. As in simulations, the change in SiO_2 thickness causes a color change. Also, the upper silver layer affects the wavelength and even more to the reflectance of the surface. The measured results seem to match the simulated ones. However, the reflectance and absorptance peaks seem to be redshifted comparing to the simulations. This hints that the layer thicknesses in the samples might not match entirely with the thicknesses used in the simulations. Because the lines in Figure 5.3 have moved towards bigger wavelengths, the thicknesses in the samples are probably a little different than in the simulations. Our guess is the top Ag layer thickness is less in real samples as compared to simulations. Because while transferring Ag to PDMS, it is not possible to transfer exactly the same thickness of the layer.

6 CONCLUSION

In this thesis, we have studied the optical properties of metal-insulator-metal structures and presented how they can be utilized as a reusable optical encryptor for security purposes. The function of MIM structures as an optical cavity has been theoretically explained and the designed structure has been presented in Figure 2.3. The structure has been studied via simulations with Lumerical FDTD Solutions software and actual samples have been made in the laboratory. The results of these experiments can be used to develop more advanced MIM surfaces and optical security methods.

According to the simulations, the thickness of the silicon dioxide and upper silver layer does have an effect on the reflectance of the MIM structure. By thickening the silicon dioxide layer, we can witness an increase in wavelength. This increase, also known as a redshift, means that the color of the surface changes. The simulations show that with different SiO_2 layer thicknesses we get a different surface color. The upper silver layer also seems to affect the surface's color and the amount of light reflected. By increasing the thickness of the upper silver layer, the MIM structure absorbs more light. This has an effect on the color and also on the contrast. With simulations, we come to the conclusion that by making small changes to the thickness, it is possible to create a pattern to the surface.

By making physical samples, we can see the MIM structure working as an optical cavity. Out of all the samples created in the laboratory, two variations produce good visible results. Both of these structures have a 10 nm upper silver layer. The silicon dioxide layer variates from 120 nm to 130 nm. By having different SiO_2 layers, the surfaces have different colors. Both of the patterns in SiO_2 are visible to the naked eye and camera. As suggested in the theoretical chapter, the patterns can be made invisible by removing the upper silver layer. Here we used the polydimethylsiloxane layer to remove the silver in a controlled matter. The silver layer can also be attached and removed multiple times, making the structure reusable.

By measuring the reflectance of the physical sample, we get a similar type of results as with the simulations. The redshift happens when increasing the thickness of silver or the silicon dioxide layer, resulting in the expected color change. However, with the samples, the reflectance peaks are in different places than with the simulations. This is because the real samples might have small differences in the thicknesses of layers.

All in all, the simulations and laboratory samples show that the MIM surface is capable of encrypting and decrypting patterns. This means that the structure can be used as an optical security feature. Taking into account how long and difficult the process of making these structures is, the MIM works as an ideal security feature. The future research in mind, there are several ways to upgrade the MIM as a security feature, including the use of periodic holes or protective coatings.

REFERENCES

[1] Bakan, Gokhan et al. Reversible Decryption of Covert Nanometer-Thick Patterns in Modular Metamaterials. Optics letters 44.18 (2019), URL: https://www-osapublishing-org.libproxy.tuni.fi/ol/fulltext.cfm?uri=ol-44-18-4507&id=418720.

[2] B. Baloukas and L. Martinu, Optical Thin Films and Coatings, A. Piegari and F. Flory, eds., 2nd ed. (Woodhead Publishing, 2018), pp.633–666.

[3] Lee, Taejun et al. Plasmonic- and Dielectric-Based Structural Coloring: From Fundamentals to Practical Applications. Nano convergence 5.1 (2018), pp.1–21, URL: https://nanoconvergencejournal.springeropen.com/articles/10.1186/s40580-017-0133-y

[4] Ghiradella, Helen. Light and Color on the Wing: Structural Colors in Butterflies and Moths. Applied optics (2004) 30.24 (1991), pp.3492–3500, URL: https://www-osapublishing-org.libproxy.tuni.fi/ao/fulltext.cfm?uri=ao-30-24-3492&id=38917

[5] Venghaus, Herbert. Wavelength Filters in Fibre Optics. 1st ed. 2006. Berlin, Germany: Springer, 2006.

[6] Nanfang Yu et al. Light Propagation with Phase Discontinuities: Generalized Laws of Reflection and Refraction. Science (American Association for the Advancement of Science) 334.6054 (2011), pp.333–337, URL: https://science-sciencemag-org.libproxy.tuni.fi/content/334/6054/333

[7] Ordinario, David et al. Stretchable Structural Color Filters Based on a Metal–Insulator–Metal Structure. Advanced optical materials 6.22 (2018).

[8] Zheng, Bo et al. Identifying Key Factors Towards Highly Reflective Silver Coatings. Advances in materials science and engineering 2017 (2017), pp.1–12, URL: https://go-gale-com.libproxy.tuni.fi/ps/i.do?p=AONE&u=tampere&id=GALE|A537719653&v=2.1&it=r

[9] Cheng, Fei et al. Structural Color Printing Based on Plasmonic Metasurfaces of Perfect Light Absorption. Scientific reports 5.1 (2015), URL: https://www-proquest-com.libproxy.tuni.fi/docview/1899508863?accountid=14242&pq-origsite=primo

[10] Chen, Qin, and David R. S Cumming. High Transmission and Low Color Cross-Talk Plasmonic Color Filters Using Triangular-Lattice Hole Arrays in Aluminum Films. Optics express 18.13 (2010), pp.14056–14062, URL: https://www.osa-publishing.org/oe/fulltext.cfm?uri=oe-18-13-14056&id=202272

[11] Inoue, Daisuke et al. Polarization Independent Visible Color Filter Comprising an Aluminum Film with Surface-Plasmon Enhanced Transmission through a Sub-wavelength Array of Holes. Applied physics letters 98.9 (2011), URL: https://aip-scitation-org.libproxy.tuni.fi/doi/full/10.1063/1.3560467

[12] Palik, Edward D. Handbook of Optical Constants of Solids. Orlando: Academic Press, 1985.

[13] CRC Handbook of Chemistry and Physics. Electronic ed. Boca Raton, Fla: CRC Press, 1978.

[14] Johnson, P B, and R W Christy. Optical Constants of the Noble Metals. Phys. Rev. B, Solid State 6.12 (1972), pp.4370–4379.

[15] Wan, Alwin M.D, Deepika Devadas, and Edmond W.K Young. Recycled Polymethylmethacrylate (PMMA) Microfluidic Devices. Sensors and actuators. B, Chemical 253 (2017), pp.738–744, URL: https://www-sciencedirect-com.lib-proxy.tuni.fi/science/article/pii/S0925400517312182?via%3Dihub