A Study Of Surface-Plasmon Polariton Dynamics In Plasmonic Quasicrystals

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I. INTRODUCTION

In all its complexity light interacts with matter fundamentally, in three ways: Transmission, Absorption and Reflection/Scattering. At a macroscopic level these lead to myriads of interesting phenomena all around us. But apparently invisible to the human eye, light-matter interactions in the nanoscopic scale, at a molecular level have generated a lot of interest among researchers. The ability to manipulate light at the sub-wavelength scale has found some diverse applications of great importance such as harvesting the solar energy to solve the energy problem [1] and in development of plasmonic biosensors based point of care devices (POCs) for biomedical research and better healthcare [2, 3],?. Fundamental research has led to discovery of some interesting phenomena such as optical trapping[?], where the light interacts with matter either by mechanical coupling leading to cooling of atoms, ions and molecules or by momentum transfer in the microscopic scale, leading to development of tools that could be used for manipulation at the cellular level. Roop Mallik et al have developed an invivo method to measure the force that is generated on transport of single organelles in a cell extract using optical trapping [4]. Another active field of research in Nanophotonics is that of metamaterials, where materials with desired electromagnetic response are being fabricated and have found novel applications in building cloaking devices, seismic detectors etc.

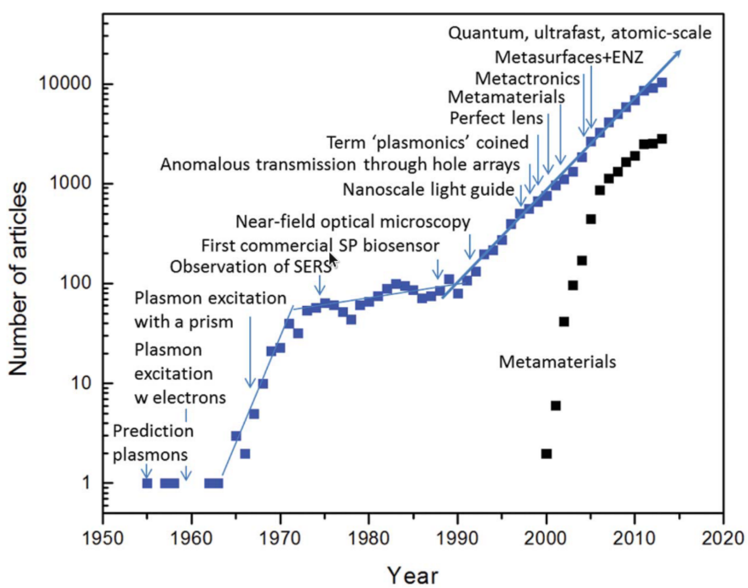


FIG. 1: Illustrations showing the surge in publications and future predictions [1].

Among these rapidly developing sub-fields in Nanophotonics are the areas of plasmonics and sub-wavelength optics. The optical transmission through thin metal films is known to be poor but, the observation of enhanced optical transmission through sub-wavelength holes due to coupling with surface modes in the metal has attracted a lot of interest. The optical properties of such air-hole arrays on a metal-dielectric interface have been studied in many different configurations and arrangements. One such system is the Plasmonic quasicrystal that has unique property of tunable broadband optical transmission. In this report we attempt to understand the underlying mechanism of coupling of light with surface plasmon modes in the quasi-periodic air hole array and also look at prospective methods that could be used for studying dynamics of surface plasmon polaritons in nano photonic structures. As the science behind it could help us develop novel applications and improvise on the already existing ones.

II. DYNAMICS OF SURFACE PLASMON POLARITONS IN PLASMONIC QUASICRYSTALS

The Concept of a "Surface Plasmon"

In condensed matter physics, free electrons in a metal are treated as a high density liquid of electrons (Fermi-Dirac gas) with a positively charged lattice as the background. Any fluctuations or oscillations in such a dense electron liquid would propagate through the bulk of the metal and such fluctuations are called "Bulk Plasmons". An important extension to this is the concept of a "Surface Plasmon" [5]. The above model was proposed by Pines and Bohm. To explain the energy losses of fast electrons passing through thin metal films, Ritche postulated a general model using hydrodynamic equations of Bloch describing the behavior of conducting electrons. According

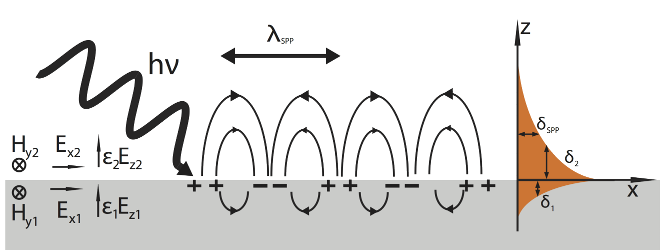


FIG. 2: Schematic diagram showing the propagation of Surface Plasmon Polariton at metal-dielectric interface.

to this model, thin metal films support a new mode of plasma oscillations in metal, the surface plasmon [6, 7]. These electromagnetic surface waves can propagate along the thin metallic surfaces or films with frequencies up to ω_p(plasmon frequency) depending on the wave vector $\vec{k}$, obeying the dispersion relation $\omega_p(\vec{k})$. A hybrid light-matter state, called "Surface Plasmon Polariton (SPP)" is formed when a surface plasmon strongly couples with light (EM radiation) of a frequency comparable to that of Ritchie frequency (ω_{sp}) [8]. Figure(2) pictorially depicts the phenomenon of coupling of a surface plasmon with light to form a surface plasmon polariton (SPP). This excitation of surface charge fluctuations (Surface Plasmons) with photons occur at metal-dielectric interface and is governed by the equations:

$$\omega_{sp} = \frac{\sqrt{\omega_p}}{1+\epsilon} \tag{1}$$

$$k_{SPP}(\omega) = k_o\sqrt{\frac{\epsilon_m(\omega)\epsilon_d(\omega)}{\epsilon_m(\omega)+\epsilon_d(\omega)}} \tag{2}$$

But, for the excitation of these surface plasmons at the interface, the primary requirement of wave vector ($\vec{k}$) matching could be achieved by using a coupling prism, scattering of light from defects and using optical nanostructures at metal-dielectric interface. There are many variations of the above mentioned methods, each with its advantages. Kretschmann and Otto configurations utilize a coupling prism [9–11]. This coupling can also occur in case of corrugated surfaces or a diffraction grating. When light is incident on the diffraction grating at a certain angle θ, the component of the diffracted light along the surface couples with the surface plasmons. The observed minima in reflectance serves as an evidence to prove that coupling occurs and a certain component of diffraction is missing.

The same phenomenon was observed by R.W. Wood in 1902 in the form of missing orders of diffraction in a metallic grating [12]. It was also observed that these anomalies were observed only in the case of p-polarized light [13]. This leads us to the important criterion of polarization dependence for the excitation of a surface plasmon. For an interface, as in the case of figure 1 where $\epsilon_1 > \epsilon_2$ there will not exist a surface mode of TE (Transverse Electric) polarization. Surface plasmon polaritons only exist with TM (Transverse Magnetic) polarization [9]. The above conclusion can be understood by solving Maxwell's equation for EM radiation propagating across a metal-dielectric interface (Maier, 2007). The concept of a "Plasmon" and especially that of a "Surface Plasmon" has been instrumental in explaining some of the fundamental interaction of light with metal nanoparticle systems and nanostructured metal-dielectric interfaces [14, 15].

Extraordinary Optical Transmission

'Camera obscura' is an age-old example, where an inverted image is obtained using a pinhole in a screen. A pinhole is an example of a simple optical element that has been fascinating researchers for centuries. There are a wide variety of applications today where the hand of a pinhole is seen. Fundamentally, the transmission of light through tiny apertures has been of great interest for its peculiar behavior. The classical theory proposed by Bethe predicts that the diffracted light from these apertures is of lower intensity than that of the incident light [16]. With rapid advances in techniques like FIB (focused ion beam lithography) and e-beam lithography in the late 1980's, it was possible to fabricate optical structures with nanometer resolution on opaque thin metal films. This led to major breakthroughs in nanophotonics and the serendipitous discovery of "Extraordinary Optical Transmission" (EOT) of light through an array of sub-wavelength scale apertures by Ebbesen et al [17].

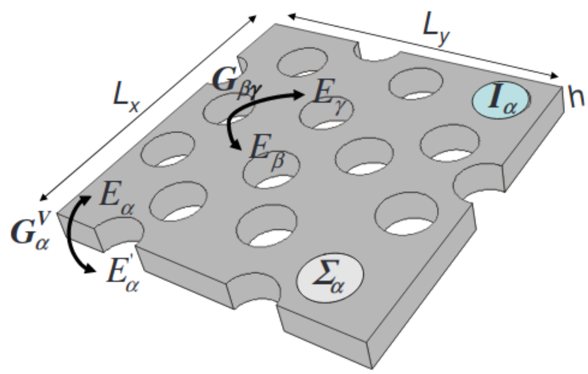


FIG. 3: L_x and L_y are dimensions of the 2D air-hole array, h is the thickness of the metal film. G_α^V represents the coupling of EM fields on both sides of the hole, $G_{\beta\gamma}$ represents the coupling between β and γ regions.

Image source: Rev. Mod. Phys. 82, 729 (2010) [18]

Their findings suggested that the transmission of light through such an array of holes on a thin metallic film was more than that of a macroscopic aperture with the same area as the sum of all the sub-wavelength apertures. Further experiments to understand the nature of the transmission through a single subwavelength scale aperture suggested a polarization dependence and requirement of metallic thin film [17, 19], clearly indicating the involvement of SPP. The

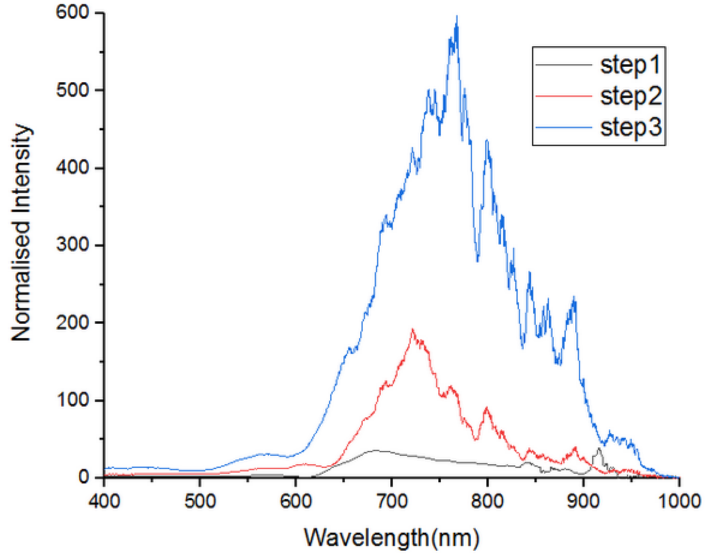


FIG. 4: Transmission Spectrum of base lattices perforated on thin gold(Au) films of 100nm thickness(Hole size = 90nm, Period = 600nm) with different rotational symmetry at normal incidence.

theoretical model proposed by Popov et al analyses the diffraction by a single aperture on a thin metallic sheet using Maxwell's equations formulated on a Fourier-Bessel basis [20]. According to their model, EOT has contribution from plasmonic excitations at the edges of the hole that are normal to the incident radiation and a weaker contribution from the electric dipoles formed at the same point due to charge accumulation [20, 21]. A number of subsequent studies were carried out by varying parameters like thickness of the metal film, hole size, symmetry of the hole arrays and different systems that satisfied phase matching conditions. The momentum conservation relation or the phase matching condition for the grating is given by:

$$k_{sp} = k_x \pm nG_x \pm mG_y \tag{3}$$

Where $k_x = (2\pi/\lambda)\sin(\theta)$ and G_x & G_y are the gratings' momentum vectors [19]. The different features seen in the spectrum (Fig. (4)) are due to coupling of discrete plasmonic modes of apertures within vicinity of each other. It follows that the position of peaks in the spectrum shift with changes in arrangement of apertures in the 2D lattice.

Therefore, it follows that with insights into the coupling one could tailor a tunable optical response of the air hole arrays for real life applications. The peculiarity of EOT phenomenon is that the transmission remains the same whether the light is incident from the dielectric side or the metal side [18]. This could be explained by the nature of coupling of multiple modes that exist at the two interfaces, air-metal and metal-dielectric interfaces. For a 2D array of holes the wave vector (k) dependence on the azimuthal angle and the rotational symmetry of the arrangement could be given by [22, 23] :

$$k_{SPP} = \sqrt{(k_o \sin(\theta)\cos(\phi) \pm \frac{2\pi n}{a_1})^2 + (k_o \sin(\theta)\sin(\phi) \pm \frac{2\pi m}{a_2})^2} \quad (4)$$

Where ϕ is the azimuthal angle and $\theta = 360/n$ for n-fold rotationally symmetric air hole array. Periodic array with crystallographically allowed n-fold symmetry is referred to as base lattice in this study. The dependence of EOT on metal has been extensively studied and reports suggest that Ag, Cu, Au, Co, Cr, Ni & W sustain EOT peaks but the first three are known to be 'Good metals' while Aluminum is considered a 'bad metal' as there is an increase in absorption. Reports have emerged showing EOT with various materials like VO_2, metal coated VO_2 double layers, highly doped Si, metal-organic conducting polymers, $SrTiO_3$ and GaAs [18]. Extensive studies reveal that the parameters that affect the EOT peaks are skin depth and absorption length. Periodic arrays of sub-wavelength holes on a metal thin film at metal-dielectric interface have been extensively investigated by Ebbesen and his co-investigators. Additionally, coupling in photonic band gap materials have also been studied. Aperiodic system and quasi periodic systems have been studied and have been found to show peculiar transmission properties on account of the arrangement of the apertures[22–24], that further validates the idea of achieving desired response for opto-electronic device applications such as optical resonant antennas, novel kind of lenses and multiplexing devices [18].

Plasmonic Quasicrystals with broadband optical response

Quasicrystals

Order, periodicity and rotational symmetry are the parameters to define crystalline forms of matter. According to the crystallographic restriction theorem, the rotational symmetries that are allowed in crystals are limited to 2-fold, 3-fold, 4-fold and 6-fold. In their seminal paper published in 1984, Shechtman et al reported the existence of long-range orientational order with icosahedral point group symmetry and no translational symmetry in a rapidly cooled alloy of Aluminum and

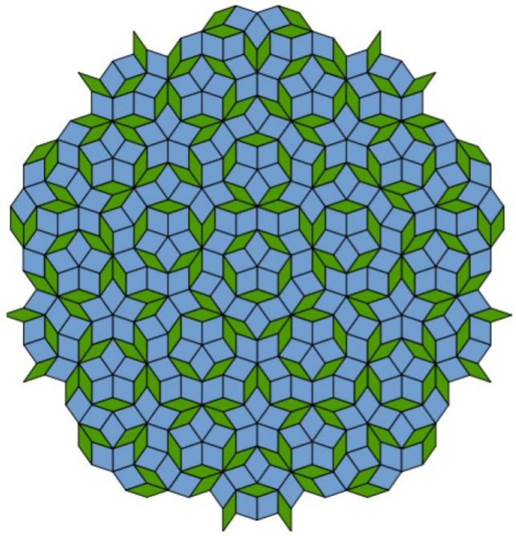


FIG. 5: Example of Penrose tiling with 'kites and darts'

Manganese [25]. This revolutionary discovery won Daniel Shechtman the Nobel Prize in the year 2011. Steinhardt et al defined an 'Ideal Quasicrystal' to be an infinite repetition of two or more distinct repeating units in space that are packed into a lattice with long-range quasi periodic translational order and long range translational order [26, 27]. This analog of a lattice unit cell is called a 'quasi lattice'.

After the discovery of quasicrystals by Daniel Shechtman, attempts were made to understand the quasiperiodic arrangement of lattice units. Unknown to crystallographers, a mathematical concept of tiling was given by Roger Penrose in 1974. Penrose tiling explained the quasi-periodicity and the kind of arrangements of basic structural units that would lead to such packing [28]. This concept of quasi periodicity is predicted to have far reaching implications in the fields of Mathematics, Physics and Chemistry. Freeman Dyson, even proclaimed that quasi crystals hold the secret to the Riemann hypothesis, one of the unsolved problems in pure mathematics [29]. Definitely, with such far reaching implications quasi periodicity has also found its way into Extraordinary Optical Transmission. Kasture et al have shown that a broadband optical transmission response is obtained with a quasi-periodic air hole array at metal-dielectric interface. To understand the origin of such features in the far field due to coupling of surface plasmonic modes is the subject matter of this study [22].

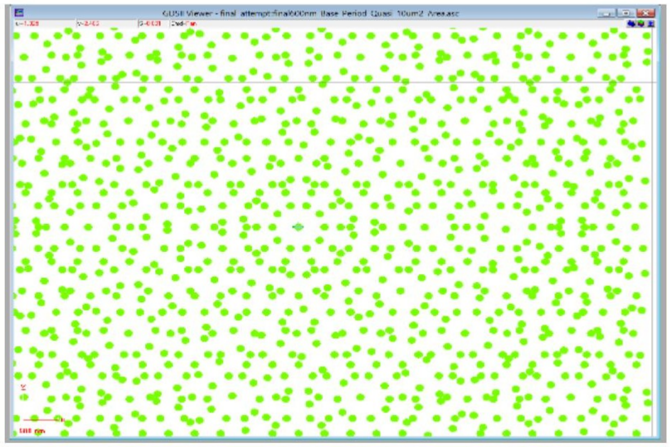


FIG. 6: Screenshot of CAD design of the complete quasicrystal

Design of Plasmonic Quasicrystals by Moiré Patterning

The patterns as in figure (1) can be made by the tiling method that involves thick and thin rhombi, one of the methods of Penrose tiling. Another alternative way of doing the same is using kites and darts. The design of 2D plasmonic quasicrystals, could be achieved in a number of ways and a detailed report of the available has been summarized elsewhere [30]. Kasture et al have used a modified Moiré patterning method to fabricate a quasi-periodic air hole arrangement [22]. They have replaced the parallel lines in the oblique tiling method with dots. In the oblique tiling method n sets of parallel lines are superimposed, each set rotated by π/n degrees before superimposition. This generates a quasi-periodic pattern of holes as illustrated in figures (6,7).

The pattern was generated by obtaining the coordinates of points for each step with necessary input parameters using a C++ program with a quick sort algorithm. The same code used in the previous reports [22] for generating the coordinates for dot exposure method of fabrication using e-beam lithography was modified to get circular holes of desired radius(r) and to sort the holes that were closer than 20-30 nm to avoid merging while fabrication of nanostructure through e-beam lithography. A similar sort algorithm with python using brute force method takes 8-9 times the time taken by the C++ code. This is due to the fact that computation is slightly intensive as each iteration involves around 14-16 million points for an area of 1600-2500 μm^2 to be patterned. Therefore, C++ yielded better results for generating the ASCII file of coordinates to generate the computer aided designs (CAD).

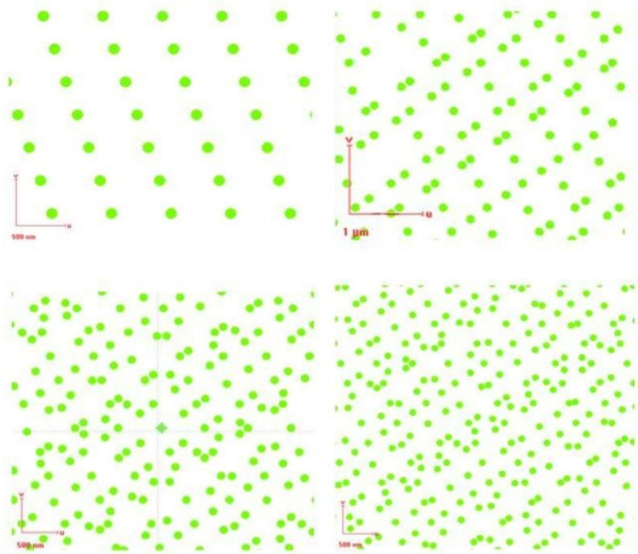


FIG. 7: CAD Images of the co-ordinates generated from the C++ program.

For the purpose of the present study, to understand the origin of the broadband response from quasi periodic patterns, we have generated the lattice coordinates for each of the 4 intermediate patterns involved in quasiperiodic pattern generation. The base lattice is a 2d lattice with the lattice oriented at $\pi/5$ angle with respect to the normal. We studied the angle and polarization resolved transmission spectra from the 5 patterns and in addition studied a randomly distributed hole pattern. In order to understand the role of plasmonic quasicrystal on the carrier dynamics, carrier dynamics when quantum dots are placed on top of plasmonic quasicrystal are studied. In the following the transmission and 2-colour pump-probe measurements will be presented. In Section 3, the complexity in sample preparation and how the problems were overcome will be presented.

III. FABRICATION AND CHARACTERIZATION OF PLASMONIC QUASICRYSTALS

Cleaning of Substrates and Sputtering Gold films

For fabricating the desired optical nanostructures on metal dielectric interfaces, quartz or silicon dioxide wafers of 300 to 500 μm thickness were taken and cut into slides of desired dimensions. For cleaning the substrate, the wafers were dipped in a beaker containing TCE (Trichloroethylene) and sonicated in a sonicator bath at 35 ^{0}C for 10 minutes, the same was repeated with acetone and methanol. The wafers were dried by blowing a current of N_2 gas. The dried wafers were then cured on a hot plate for 2 minutes at around 200 ^{0}C and cooled to room temperature.

Gold films of 100 *nm* thickness were deposited on the substrates by DC sputtering. The vacuum chamber is depressurized and the cleaned substrates are placed inside the sputtering chamber and valves are sealed. A base pressure of 4×10^{-6} mbar is set and later the pressure within the chamber was then brought to a sputter pressure of 8×10^{-3} mbar. Once the base pressure is attained, with the aid of a Mass Flow Control (MFC) system the flow rate of Argon (Ar) gas was set at 25 sccm (standard cubic centimeters per minute). A DC voltage of 0.42 kV was applied across the electrodes with a current of 0.12 amperes for 100 seconds to obtain a gold (Au) thin film of 100 *nm* thickness. The above parameters were previously optimized for generating stable plasma to sputter the metal target and coat the substrate. After sputtering is done, the DC is turned off and the chamber is depressurized to get the sputtered samples out. The sputtered samples were used for further studies involving fabrication of optical nanostructures.

Fabricating the Quasi-periodic pattern onto the Gold film

On the sputter coated substrates a layer of positive e-beam resist PMMA (Polymethyl methacrylate) was spin-coated at speed of 2000 rpm for 45 seconds with a ramp of 400 rpm/s. The spin-coated substrate was baked over a hot plate for 2 minutes at 170 ^{0}C and cooled to room temperature.The patterns of the five base lattices with rotational symmetries $\pi/2$, $\pi/4$, $\pi/3$, $\pi/5$ & $2\pi/5$ and that of each step of the oblique tiling method to fabricate the plasmonic quasicrystal were patterned on the polymer(e-beam resist) thin film by performing Electron beam lithography (EBL) using Raith e-line. Since most patterns contained a high density of circular apertures to be patterned, dosage scaling in e-line was used to optimize the right dosage to avoid merging of two or more holes close to each other. The patterned PMMA was further developed in a mixture of MIBK: IPA (Methyl isobutyl ketone: isopropyl alcohol) taken in a ratio 1:3 for 90 seconds followed by IPA treatment for 60 seconds. The developed samples were etched using RIE (Reactive Ion Etching) to Argon sputter the exposed regions of gold. The etching was done with an RF (radio frequency) power of 138 W and chamber pressure was maintained at 0.5 Pa, the mass flow rate of Ar being 50 sccm. The etch rate was previously optimized to be 10 nm/min for the above given parameters. For etching the 100 nm gold films, a 2-minute break was given after every 1 minute of etching to prevent the polymer resist from hard baking. The e-beam resist was stripped off the substrate surface using O_2 plasma at 80 W RF power for 10 minutes with 1-2 etch stops to prevent any damage to the gold surface.

Characterisation

The fabricated samples were characterized by using nanoscale surface imaging techniques such as Scanning electron microscopy (SEM) and Atomic force microscopy (AFM). The SEM images in figure() show step by step evolution of a Plasmonic Quasicrystal fabricated on a metal thin film by the modified oblique tiling method. Additionally, atomic force microscopy(AFM) could be used to determine the surface features of these nanoscale circular apertures.

IV. EXPERIMENTAL STUDIES

Angle-Resolved Optical Transmission studies

Optical transmission studies were carried out using the setup illustrated in figure (8). A 100W tungsten halogen lamp was used as the light source in the experiment. The collimated beam was focused on the sample through an objective and the transmitted beam was collected using Ocean FX spectrometer with wavelength scan range being 200-1100 nm and an integration time of 100ms. For the angle-resolved transmission measurements a movable rotating stage with a resolution of 0.03^0 was used to rotate the sample and take transmission measurements in steps of 0.3^0 from -12^0 to $+12^0$. A Glan-Thompson polarizer was used to get polarization dependent studies. The explanation to the various dispersion modes in the base lattices could be understood using the dispersion relation:

$$k_{SPP} + k_x = G^{(i)} \tag{5}$$

Where $G^{(i)}$ represents the reciprocal lattice vectors of the arrangement. Periodic have both order and rotational symmetry while aperiodic systems with either quasi-periodic or random arrangements do not have the short range order.

The quasi crystals or quasi-periodic patterns have long range order but no short range order. But, their discrete Fourier transform vectors or reciprocal lattice vectors are known to unusual orders rotationally not allowed by crystallographic theorem. In a periodic system the underlying mechanism EOT involves coupling of discrete resonances. On the other hand, in case of quasi periodic arrangement the transmission enhancement might be due to a combination of different

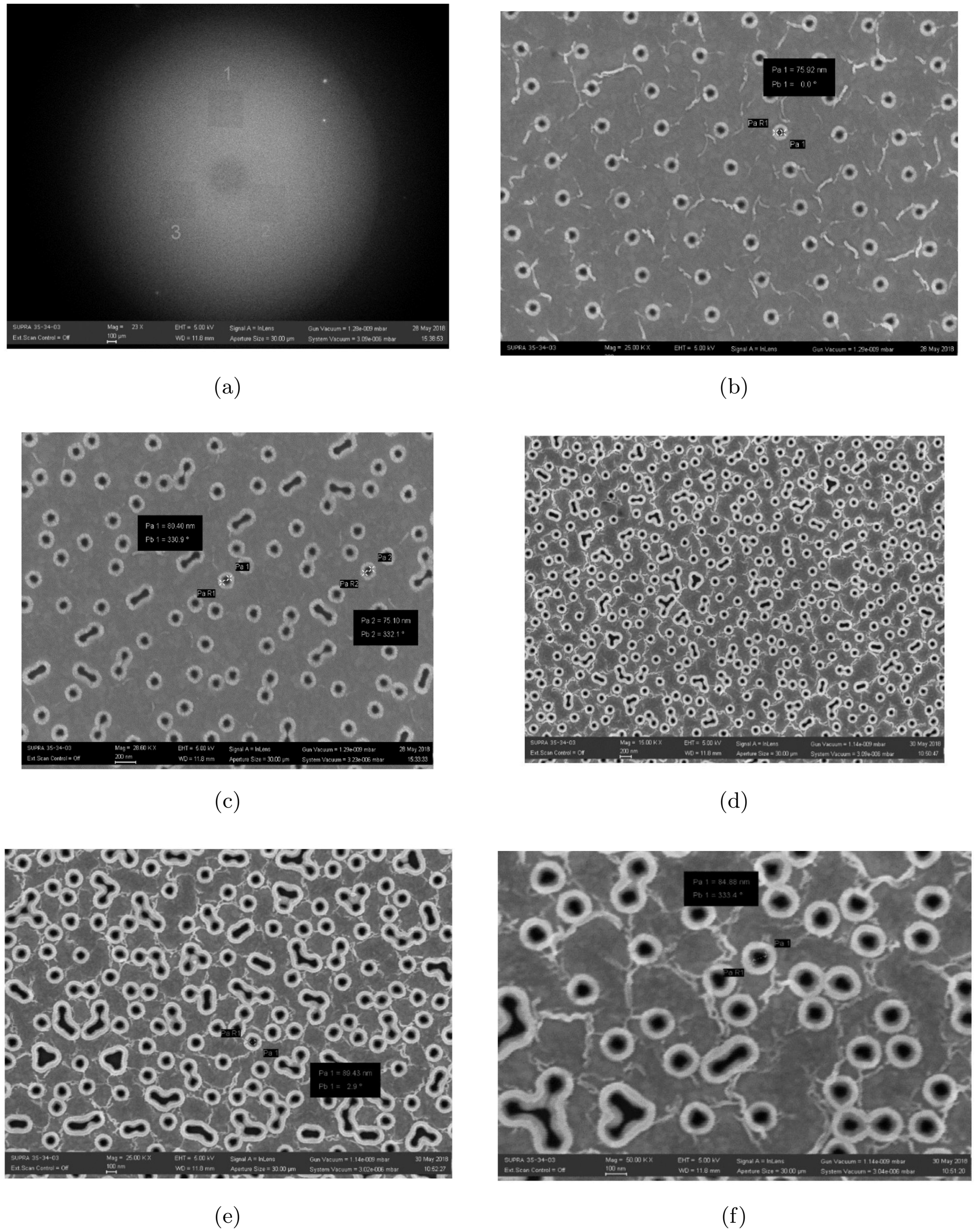


FIG. 8: SEM Images

modes. For example, the resonant continuum modes that are formed by very closely spaced apertures that are closer than 50-70 nm, the resonant discrete modes that originate due to resonance of single apertures and the interference of light transmitted from non-resonant apertures. As clearly indicated in the plots below as we go step by step increasing the holes through the oblique tiling

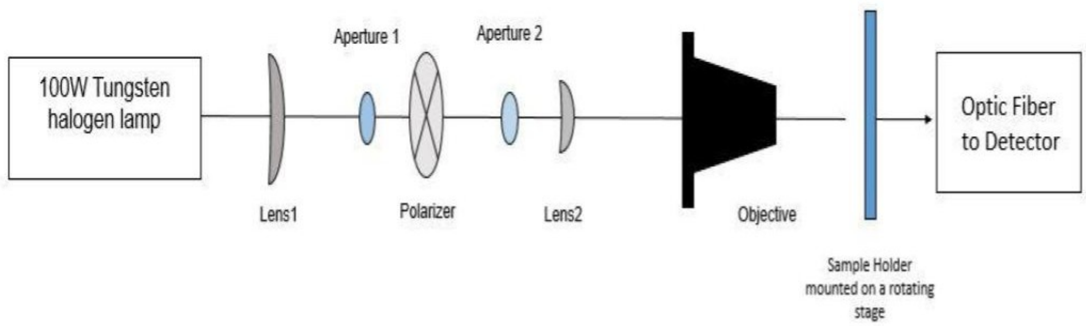


FIG. 9: Setup Scheme for Angle-resolved measurements

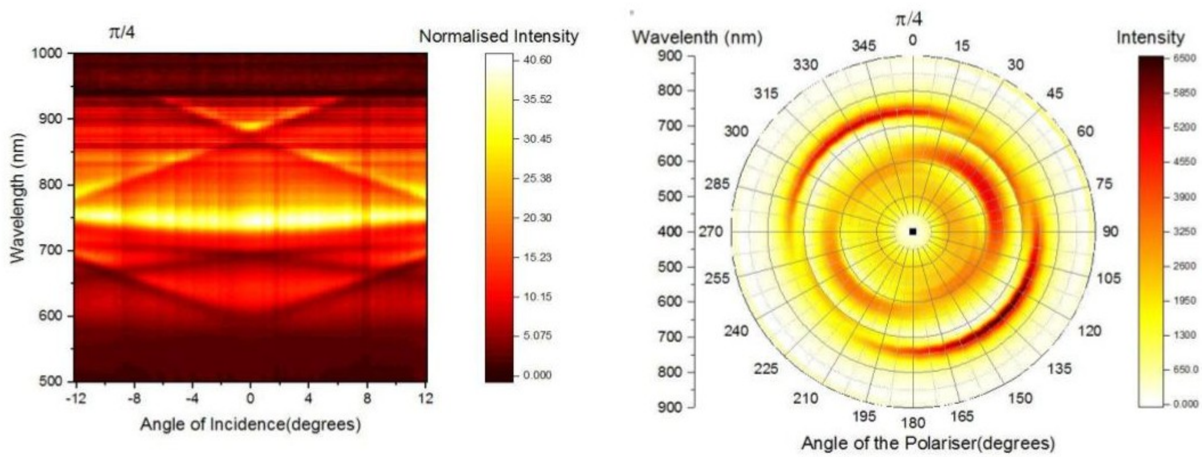


FIG. 10: Angle resolved dispersion and polarisation dependence plots for $\pi/\mathbf{4}$ base lattice

method the number of continuum modes increases and as a result of rotational symmetry the surface plasmon modes are spread equally across the patterned area. This results in a slow loss of polarization dependence of transmission enhancement of the patterned arrangement which is evident from the plots in figures (14b, 15b &16b).

Pump-probe spectroscopy to understand charge carrier dynamics

Time-resolved spectroscopic methods provide ways to understand the dynamics of processes which occur in the timescales of a few picoseconds to femtoseconds. There are multiple methods

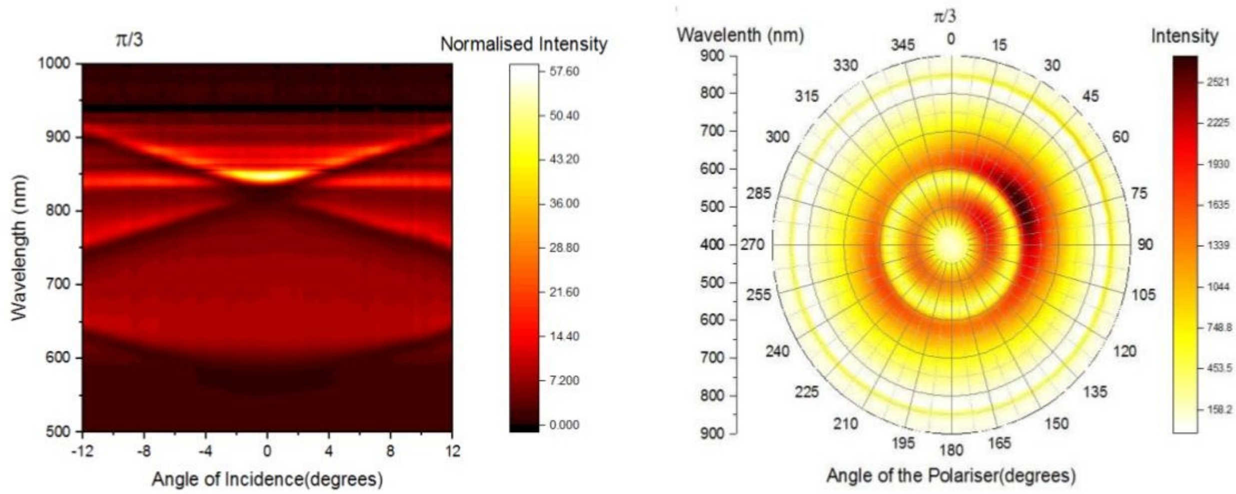


FIG. 11: Angle resolved dispersion and polarisation dependence plots for $\pi/\mathbf{3}$ base lattice

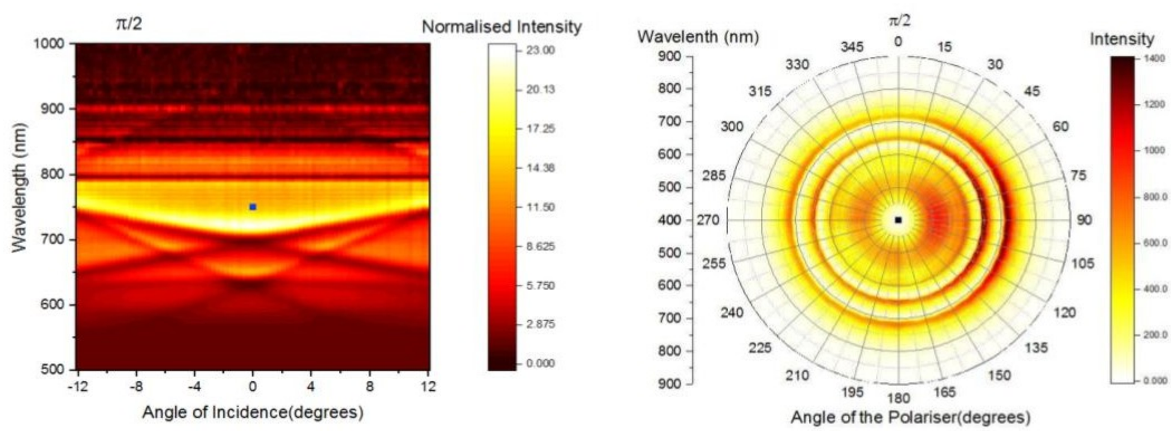


FIG. 12: Angle resolved dispersion and polarisation dependence plots for $\pi/\mathbf{2}$ base lattice

to probe these ultrafast processes, some of them like Four Wave mixing (FWM) that involves intermodulation based on non-linear optics. A more general and broadly used technique is the tunable multiple wavelength "Pump-Probe Spectroscopy". This technique involves the use of two ultra-short pulses. A pump pulse to excite the sample, to generate a non-equilibrium excited state and a probe pulse to measure the changes in the excited state. A degenerate Pump-probe configuration was setup using a Ti: Sapphire femtosecond (fs) pulsed laser (Mai Tai, Spectra Physics) with Spectra Physics' Solstice Ace ultrafast amplifier and TOPAS (the automated optical parametric amplifiers).The above combination allows a wide tunability of wavelength, ideal for

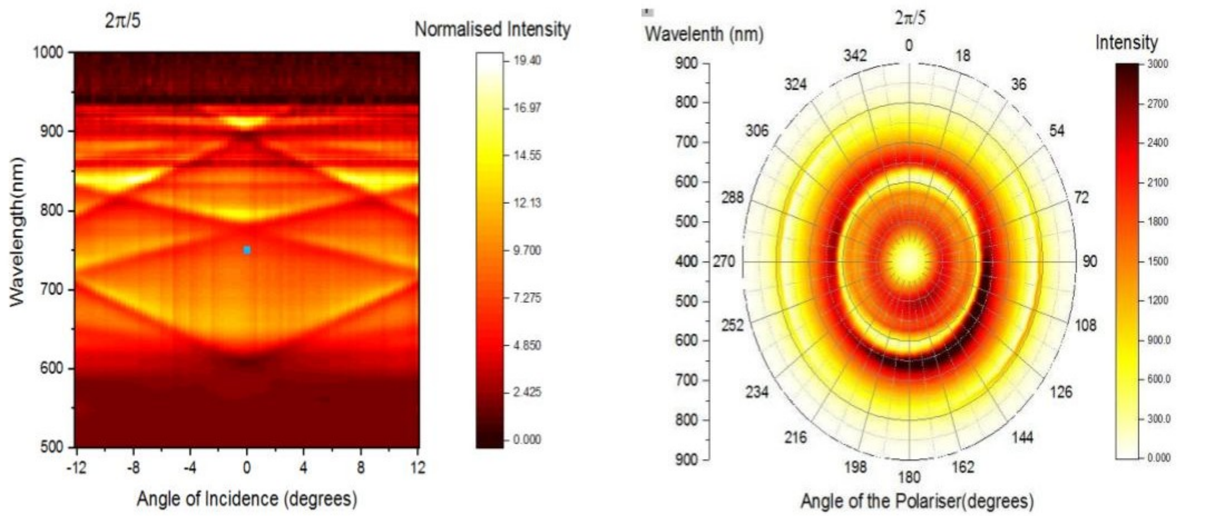


FIG. 13: Angle resolved dispersion and polarisation dependence plots for $\mathbf{2}\pi/\mathbf{5}$ base lattice

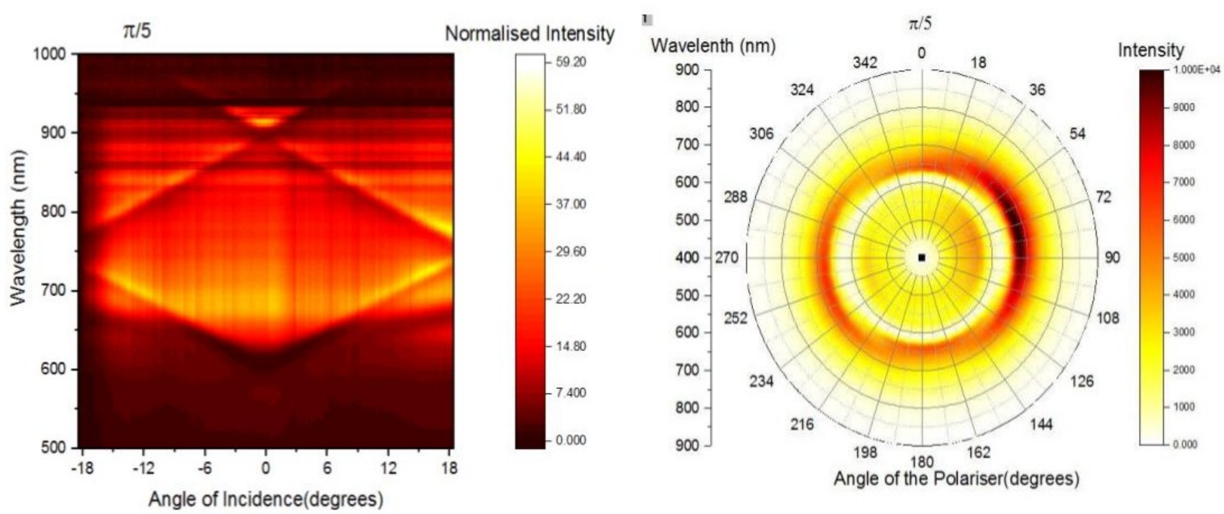


FIG. 14: Angle resolved dispersion and polarisation dependence plots for $\pi/\mathbf{5}$ base lattice

a multicolor pump-probe experiment. The pulse width obtained was (FWHM) 35fs with the repetition rate being 1 KHz, a low repetition meant that there would be no contribution from thermal excitations of the electrons in the metal thin film. The output beam from the solstice ace at 800 nm was taken as the probe and the output beam from TOPAS tuned to 500nm was taken as the pump [31–33].

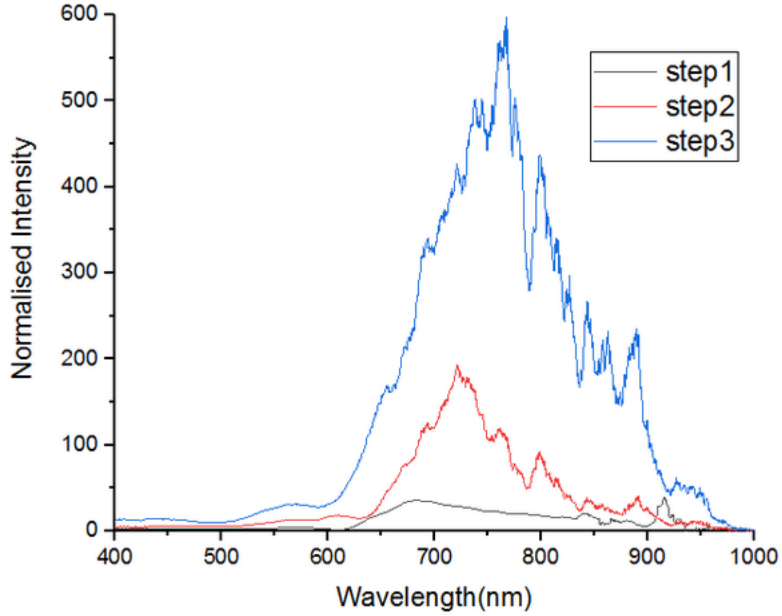


FIG. 15: Transmission Spectrum of different steps (Hole size =70nm, Period = 600nm)

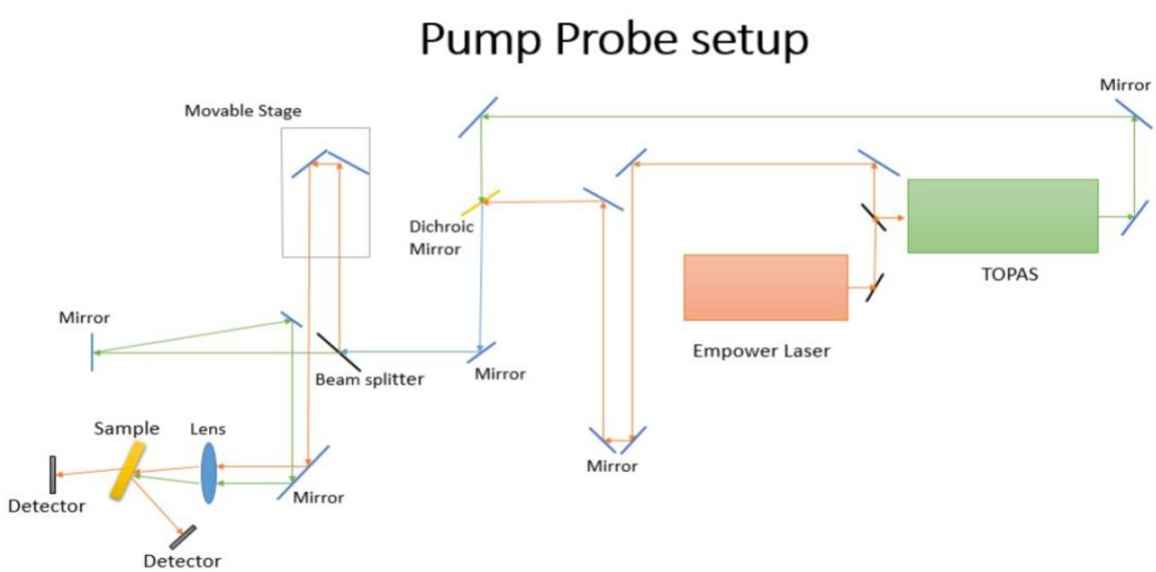
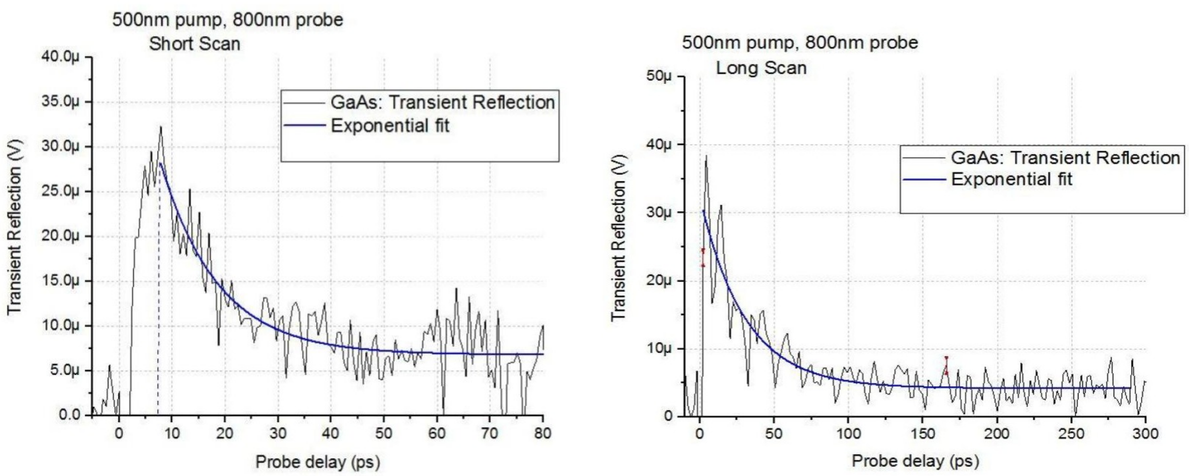


FIG. 16: Schematic diagram of the optical setup used for the Pump-probe experiments

GaAs (Gallium Arsenide) is a direct band gap semiconductor material with high charge carrier mobility that has high reflectivity uncommon in semiconductors. By virtue of such properties GaAs has found wide range applications in optoelectronics with development of bulk GaAs and GaAs



(a) Short Scan (b) Long Scan

FIG. 17: Lifetime measurements of charge carriers in GaAs samples to benchmark the pump-probe setup.

quantum wells. With pump fluence being 0.5 J/cm^2 the lifetime of the charge carriers in GaAs was estimated to be 10 ps (picoseconds) in a quick short scan, while a slower long scan gave a lifetime of 26 ps.

Data Analysis

All the data collected during this project was analysed with the aid of programs written in Python using the interactive ipython notebooks in jupyter. The huge number of ASCII files generated for each samples' transmission measurement were all compiled, normalized and visualized using the libraries numpy, pandas and matplotlib in python. To generate the plots used in this report and for curve fitting, Origin 2018 was used by interfacing it with python codes through the python console feature available in Origin 2018.

V. CONCLUSION & FUTURE SCOPE

In summary, the current study has provided a basis to gain insights on the origin of the broadband optical transmission enhancement of light through plasmonic quasicrystals. The increase in density of holes that are closer to each other lead to formation of resonant modes that cause the broadening of the transmission peak and also the loss of polarization dependence.

The potential of tunable extraordinary optical transmission through structured arrangement of holes provides scope for novel applications. A theoretical model to generate simulations for transmission through quasiperiodic patterns could help us better understand the underlying phenomena and this would be an ideal goal to pursue in light of the findings of this report. With an understanding of the relationship between geometrical pattern and optical response, new structures could be designed using machine learning models with or without supervised learning algorithms. This could lead a new synthetic approach to design plasmonic crystals.

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