Corner-Promoted Focus Enhancement of Light in Conical Holes for Extraordinary Optical Transmission

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Abstract—Extraordinary optical transmission (EOT) is generated when light transmits through an array of subwavelength holes on a metallic sheet. Most studies have focused on the EOT effect with ideal cylindrical holes. Subsequently, it has been recognized that imperfection in hole fabrication could alter light-matter interaction. Later, adiabatic taper is reported to promote light nano-focusing effect in the optical and near infrared wavelength regime. Due to geometrical and fabrication constraints, it is difficult to fabricate adiabatic taper for mid-infrared responsive nanohole array. In this work, we report the construction of nanohole arrays with varying near-adiabatic taper holes. We demonstrate that when the positive (inverted conic) angle increases from 10° to 20°, the peak full width half maximum (FWHM) improves by 14% from 800 to 700nm, respectively. Furthermore, inverted conical hole is shown to be superior to negative conic angle. Unlike cylindrical hole array, conical hole array maintains high transmission throughput for any given hole dimension while achieving better FWHM. The betterment in performance for conical over cylindrical holes is attributed to a localized plasmonic effect at the terminal exit. Subsequently, the device is demonstrated as a CO2 low flow sensor. The highest sensor sensitivity is determined to be between 1–4sccm.

Index Terms—Carbon dioxide, gas sensor, nanophotonics, nanostructures, nanofabrication.

I. INTRODUCTION

Many different sensing platforms, such as plasmonic sensing, have been realized and evaluated for a multitude of applications [1]–[4]. In 1998, Ebbesen et al discovered the interesting phenomenon of light enhancement by means of perforated holes in metallic thin film [5]. The experiment demonstrated light-matter interaction between the incidental light and that of surface plasmons. The pioneering work showcased the possibility of the 0th order transmission surpassing unity. In turn, the new discovery overturned the then understanding based on Bethe’s theory of diffraction [6].

In the presence of an array of holes drilled into metal films, coupled with geometrical design, maxima and minima peaks are revealed. Maxima peaks correspond to surface plasmons resonant excitation while the minima is associated with Wood’s anomaly [7]. Today, the phenomenon is commonly described as the extraordinary optical transmission (EOT). EOT phenomenon is brought about by the surface plasmon polariton, localized surface plasmons and transmission Fabry-Perot effects [8]–[10].

Ever since then, many research works have confirmed the essentiality of geometrical parameters to manipulate the EOT transmission profile. These efforts include the alteration of period, diameter of the holes, hole shapes, unit cell arrangement, thickness of the metallic layer and the use of different metals depending on the intended transmission frequency [11]. In particular, it has been shown by theoretical and experimental works that a change in period could shift the transmission wavelength. Earlier works focused on elucidating the nature of EOT that spanned localized EOT from isolated and randomized holes placement, light-molecule interaction induced absorption transparency, Brewster angle EOT and single light hole aperture [12]. In recent years, published works are largely application-based [13], [14]. In general, EOT for sensing applications is widely reported [15]–[17]. Broadly, these molecular sensors are centered on surface plasmonic...
resonance as well as fluorescence measurement enhancement for single molecule detection. Besides that, color filtering, perfect absorber, meta lenses, optical trapping, lasers, etc have also been demonstrated [18], [19].

The myriad of applications mentioned is generally realized in the optical to the near infrared regime. Due to the sub-wavelength hole size and other stringent geometrical requirements, high precision nanolithography tools have to be used. A majority of the reported works in the literature are fabricated using either single-step focused ion beam (FIB) or electron beam lithography (EBL) followed by metal deposition and liftoff [20]–[22]. Apart from that, other fabrication techniques such as nanoimprint, nanosphere lithography, laser interference lithography, etc have been proposed as alternatives to FIB and EBL techniques [23]–[25].

Amongst the techniques, FIB, EBL and laser are typical processes. Comparing EBL, FIB and laser-based techniques, the latter is operable at non-vacuum condition. On the contrary, EBL and FIB have to be operated under high vacuum in specialized chambers, driving up fabrication and operating costs. On the other hand, EBL is more scalable for possible production while the laser and FIB approaches being more time consuming are generally suitable for small scale manufacturing and research. The main shortcoming of laser-based lithography is that the technique is hampered by its wavelength-dependent optical diffraction limit. However, recent advancements, as reported in a few publications, has highlighted the performance of laser lithography technique with the potential of challenging that of traditional FIB technique in the future [26], [27].

On the other hand, the focus of light in a single narrowing light pipe have been investigated previously [28], [29]. Such field distributions and its associated localized field enhancement can be achieved through the modification of the structural design to realize \( \text{Re} [\varepsilon_m + 2\varepsilon_d] \rightarrow 0 \) [11]. So far, tapered metal rods, glass sphere tip and tapered air gap have been reported[30] Many of those studies were carried out with adiabatic taper where the critical angle,

\[
\theta_c \approx \left| \frac{2 \varepsilon_d}{\varepsilon_m} \right| 
\]

where \( \varepsilon_d \) and \( \varepsilon_m \) are the permittivity of the dielectric medium in the taper and the real part of the metal permittivity. In addition, any design deviation from the critical taper angle would result in a decline in coupling efficiency along the length of the taper. As a consequence, large angle (\( \theta \gg \theta_c \)) results in scattering by mismatched waves. On the other hand, absorption losses dominate low coupling efficiency [31]. However, it has to be noted that the laid out adiabatic condition is a mere reference that could well underestimate the actual critical angle value [29].

So far, most studies focused on realizing a single nanotip output port [32]. The output terminal is typically much smaller than the input terminal. As such, the critical angle for adiabatic surface plasmon propagation decreases with an increase in real part permittivity of metals as a result of a decrease in SPP frequency at longer wavelength. In addition, as reported by Choo et al, the critical angle is also determined by the thickness of the metal layer [31]. Due to the difficulty in fabricating such feature for longer wavelength, conical nanohole array is rarely reported for mid infrared and longer wavelength radiation. Recently, Chen et al investigated the effect of stepped and sloped taper nanohole array by simulation [33]. Despite the increase in transmission intensity for stepped taper, its output response suffers from low quality (Q) factor.

In this work, mid infrared responsive tapered (conical) nanohole array is demonstrated. Through simulation and experiment works, an enhancement of EOT effect, in terms of transmission peak full width half maximum (FWHM) and transmission intensity, brought about by the change in nanohole taper angle will be investigated. At the same time, the fabricated sensor is validated as a low flow CO2 gas sensor.

II. EXPERIMENTAL DETAILS

Nanohole array was patterned on CaF2 substrate by FIB or EBL+liftoff to create nanoholes with different taper angles. CaF2 (100) wafers were first cleaned in acetone and isopropanol under sonication. For FIB fabrication, 50 nm of Au was deposited directly on CaF2 by electron beam evaporation. The nanohole array was then etched with Ga as source. In this case, samples of dimensions 50 \( \mu \text{m} \times 50 \mu \text{m} \) were FIB-ed by a ZEISS Crossbeam 540 FIB-SEM. The FIB probe acceleration voltage and current used for creating such array was set to 30 kV and 50 pA. Then the surface was further etched to remove the possible Ga remnants.

For EBL process, a thin coating of AR-PC5090 conductive polymer was applied. Then, A4 PMMA was spin coated onto the CaF2 substrate and processed according to recommended specifications. It was then subjected to EBL using 1400 \( \mu \text{C/cm}^2 \). The pattern was carefully developed in MBK-IPA (1:3) for 1 min. Thereafter, 5 nm Ti and 50 nm Au were evaporated in sequence. Finally, metal liftoff was carried out in acetone.

The simulation was setup as follows. The material properties of Au and Ti, used as-is from the default material database, were obtained from the CRC Handbook of Chemistry & Physics. On the other hand, CaF2 is allocated a constant refractive index of one. The period (\( P \)) was set to vary the \( \theta \) and \( \varepsilon_m \) spans to be between 2.1 and 2.9 \( \mu \text{m} \). The radius of the top hole (\( r_1 \)) was fixed at 0.625 \( \mu \text{m} \) throughout. Then, a plane-wave source was fixed at a location 15 \( \mu \text{m} \) away from the nanohole. The model mesh accuracy was set to 2 to balance between accuracy and system requirement which affects the overall runtime. To improve the simulated results accuracy, a high accuracy mesh was assigned to the nanohole with a buffer of 300 nm. Finally, the simulation was performed using Numerical finite difference time domain (FDTD) module with a simulation time of 12000 fs at 300K. The boundary conditions of \( x, y \) and \( z \) were set to anti-symmetric, symmetric and PML, respectively with an auto shutoff criterion of 1e-5.

III. RESULTS AND DISCUSSION

To verify the effects of non-vertical shape etch holes, conical shapes hole was simulated with top and bottom radii, \( r_1 \) and \( r_2 \), as shown in Figure 1.
The simulated results of the conical holes \((r_1 > r_2)\) are compared to that of perfect cylindrical holes where \(r_1 = r_2\). The latter is typical described geometry for most EOT design experiment. Although desirable, it is all but highly challenging to fabricate due to equipment and process limitations. Typically, a positive or negative slope would be obtained experimentally. From a practical perspective, it is more meaningful to simulate sloped features. Thus, a simulation was carried out to understand the effect of varying \(r_1\) and \(r_2\). As shown in Figure 2, a combination of different \(r_1\) and \(r_2\) scenarios were simulated with \(P = 2.5\, \mu m\).

It can be observed from Figure 2A that variations in \(r_1\) and \(r_2\) values with values less than 4% do not change the transmission wavelength. On the other hand, when \(r_2 < r_1\), the peak full width half maximum (FWHM) decreases. Similar enhancement is obtained when \(r_1 < r_2\). However, the FWHM enhancement (which corresponds to a smaller FWHM value) is greater for \(r_2 < r_1\) than \(r_1 < r_2\). A change from perfect cylinder to inverted conical hole array brings about an improvement in transmission Q factor. Further reduction of \(r_2\) (<4%) with fixed \(r_1\) leads to a significant decrease in transmission intensity albeit incremental improvement in FWHM.

Generally, the transmission profile of a nanohole array can be approximated by the following equation (2)

\[
\lambda_{\text{peak}} = \text{Re} \left( \frac{P}{\sqrt{j2 + j2}} \frac{\varepsilon_d \varepsilon_m}{\varepsilon_d + \varepsilon_m} \right)
\]

(2)

where \(P\) is the nanohole array period, \(i\) and \(j\) are integers, \(\varepsilon_d\) and \(\varepsilon_m\) are the relative permittivity of the dielectric and metal layers. A change in period, as shown in Figure 2B, would affect the transmission passband, \(\lambda_{\text{peak}}\). In this case, smaller period corresponds to shorter \(\lambda_{\text{peak}}\) while larger period corresponds to longer \(\lambda_{\text{peak}}\). As simulated, transmission intensity decreases and a smaller FWHM at longer \(\lambda_{\text{peak}}\).

Possibly, the EOT quality can be enhanced by decreasing the hole radius while keeping the period constant. It has been shown in earlier works that a decrease in hole radius improves the transmission peak FWHM. However, a consequential decrease in transmission intensity is then expected. In this particular case, it is interesting to determine if the enhancement is a result of hole narrowing and/or tapering.

Therefore, FDTD simulations involving cylindrical and conical hole arrays of various bottom hole radius, \(r_2\), are carried out. With reference to Figure 3A, it can be observed that the transmission profiles of conical holes \((r_1 > r_2)\) are different from that of cylindrical holes. Namely, the simulated transmission intensity response of conical holes is greater than that of cylindrical holes (Inset I). It is expected that a smaller hole dimension will result in a lower transmission intensity. According to Figure 3A (Inset II), the decrease in intensity is less for conical than cylindrical nanohole arrays. Moreover, the difference is more explicit for smaller \(r_2\) radius. As illustrated in Figure 3B, a localized plasmonic effect can be induced with a change in the edge angle. With a progressive difference in radii, \(r_1\) and \(r_2\), greater localized plasmonic effect is promoted. As a result, localized plasmonic ‘hotspot’ at the exit terminal end enhances the overall electric field. Comparing the case of \(r_1 = r_2 = 0.625\, \mu m\) with that of \(r_1 = 0.625\, \mu m, r_2 = 0.55\, \mu m\) hole arrays, the field intensity enhancement of the conical system is 40 % more than that of the cylindrical system. Similar to previous reports, by using noble metal nanotip, field enhancement can be further improved with the alteration of the cone geometrical design [34].

On the other hand, with reference to \(r_1 = r_2 = 0.625\, \mu m\) and \(r_1 = r_2 = 0.550\, \mu m\), the “squeezing” effect due to a
narrower aperture diameter can also affect the transmission response, such as the FWHM. This fact illustrates that conical hole might influence the transmission profile differently as compared to cylindrical hole. Therefore, the taper angle is an additional parameter for the alteration of optical response independent of hole radius. In which, transmission profile can be finetuned in addition to hole radius and period manipulation.

Previously, the effect of Ti adhesion layer has been investigated. Ti is commonly used as an interlayer thin film to attach Au film onto an underlying substrate or thin film. In many cases, Ti of up to 10 nm is sputtered or evaporated. It has been shown that the inclusion of Ti interlayer would 1) redshift and 2) reduce the plasmonic peak attributed to its high damping frequency [35].

According FDTD simulation as shown in Fig 4, significant peak shift by 50 nm and the decrease in transmission amplitude associated with Ti interlayer are observed. The phenomenon can only be observed when the Ti thickness is more than 10 nm. On the contrary, a 5 nm Ti layer would only cause an approximate 10% decrease in transmission. It does not cause an observable redshift in peak position.

Similar to results documented in published reports, the presence of a Ti interlayer worsens the transmission peak. By using FDTD simulation, it is proven that the FWHM increases from 0.210 µm (Ti 0 nm) to 0.249 µm (Ti 10 nm) for a cylindrical hole array. Interestingly, the inclusion of a Ti interlayer improves the transmission peak FWHM for a conical hole array. For a Ti 5nm interlayer thickness, the FWHM decreases from 0.210 µm (0.625 µm – 0.625 µm) to 0.187 and 0.163 µm for (0.625 µm – 0.600 µm) and (0.625 µm – 0.580 µm) conical holes, respectively.

To demonstrate the effect experimentally, holes were fabricated by both 1) EBL followed by liftoff (denoted as EBL+liftoff) and 2) FIB. The former technique typically yields almost-vertical thin film profile after deposition and liftoff. On the contrary, like other dry etching techniques, FIB-ed structure can be moderated according to the milling process conditions. Subsequently, EBL and FIB prepared samples were subjected to transmission electron microscopy.
It can be seen in Figure 6A and B that the transmission passband is $\lambda_{\text{peak}}$ period dependent. Similar to simulation, the $\lambda_{\text{peak}}$ red shifted with larger period. Both EBL and FIB samples have similar $\lambda_{\text{peak}}$ between 3.5 and 5.0 $\mu$m. The main difference between transmission profile of EBL and FIB is that of the transmission intensity. Comparing the $\lambda_{\text{peak}}$ when $P = 2.1 \mu m$, the transmission intensity is approximately 17.5 % and 30% for EBL and FIB samples, respectively. It can be observed that the FIB peaks intensities are approximately twice that of EBL peaks intensities. The decline in transmission for EBL-made samples is attributed to the decrease in plasmonic propagation length caused by the Ti layer [38]. In addition, we report an improvement in Q factor when the hole is conically shaped. According to Figure 6C and 6D, a decrease in slope gradient of about 20° translates to a decrease in FWHM from 800 nm (EBL+liftoff) to 700 nm (FIB). The outcome is similar to simulation and previous reports such as [28].

Nanohole array can be fabricated for a multitude of EOT sensors. One such use cases is the fabrication of miniaturized spectrometers. The variation in period, as shown in Fig 2B, allows one to construct a 1D or 2D sensors. For simplicity, the FIB samples were later explored as an absorption-based CO2 gas flow sensor. The steady-state flow rate of CO2 was passed through the overhead space, between that of the sample and the light source. In this case, CO2 molecules which absorb incidental radiation at 4.26 $\mu$m, would attenuate the transmission intensity according to the volume of CO2 present. As shown in Figure 7A, CO2 gas flow is moderated between 4 and 10 sccm by a mass flow controller. The best fit curve, as plotted in Figure 7B, reveals a R-squared value of 0.996. It can be observed that the maximum sensitivity, determined by the slope of the curve at a given point, of the sensor is when the flow rate is between 0 and 4 sccm. Within the range, due to the sensitivity of the sensor, small change in flow rate could be precisely discriminated. On the other hand, the maximum detection limit is extrapolated to be approximately 23.6 sccm. This work epitomizes the use of EOT sensors as a potential high sensitivity low flow gas sensor for industrial and healthcare uses. Potential use cases include insufflation gas flow monitoring during laparoscopic surgery and CO2 level moderation in greenhouses.

While numerous mid infrared plasmonic filters have been published, their performances are rarely tabulated and compared. As shown in Figure 7C, best reported experimental FWHM from recent publications on free space plasmonic filters are included and compared. A smaller FWHM which translate to better sensor quality, is preferred. Generally, from Figure 7C, plasmonic filters FWHM increases at longer wavelength. This work showcases an improved FWHM for both EBL and FIB fabricated devices. However, reports that utilize molecule gas traps such as metal organic framework, polyethyleneimine as well as works which do not reveal the transmission / reflection profiles are excluded. As compared to other state of the art sensors, the developed sensor’s quality factor (FWHM) at 4.26$\mu$m is at least 2.4 times higher.

The enhancements in FIB transmission intensity and FWHM are further investigated. Recently, gallium has been
Fig. 6. Experimentally obtained transmission period specific passbands for A) EBL and B) FIB samples with periods between 2.1 and 2.9 μm. Comparison of passband for period 2.5 μm for C) EBL and D) FIB showing a 14% enhancement of FWHM for the latter.

reported to display plasmonic effect [46]. Thus, one of the main questions is whether the presence of a thin layer of Ga contamination introduced during the milling process is responsible for the enhancement [47]. Firstly, area scanned TEM energy dispersion spectroscopy (TEM-EDX) was carried on EBL-liftoff and FIB samples as shown in Figure 8. For EBL sample, the presence of Ca, F and Au and the absence of Ga as marked by the red circle suggest the following. 1) FIB milling with Ga source used during surface investigation does not contribute significantly to surface Ga contamination. 2) Serve as a confirmation reference for FIB sample. In contrast, the FIB sample, as shown in Figure 8B, reveals a detectable amount of Ga as detected by TEM-EDX. Therefore, surface contamination by Ga must be accounted for in processes that involve Ga ion milling. As such, alternatives such as noble gas milling have been developed. Although it can eliminate Ga contamination, one drawback is that the process is time-consuming and uneconomical for large area hole milling operation.

Secondary to ion milling, FIB sample was further processed to remove the deposited Ga. In this case, a low acceleration voltage (KV) at 5 kV and current at 10 pA etching step was used. The main purpose of this step is to remove the implanted Ga by means of a physical process. The setup of the steps is as follows. As shown in Figure 9, the nanohole array was divided into four quadrants (I – IV). Thereafter, the deposited Au film on the bottom half of the hole array (III, IV) was gently scrapped off using a sharp tip tweezer. The left-half of the sample (I, III) was then subjected to low KV cleaning. The quadrants were then optically measured. The aperture size was set to be 12.5 μm x 12.5 μm. As shown in Figure 9B, the transmission between quadrants (I, II) were compared. Marginal change to FWHM and transmission intensity could be observed. Both Ga only quadrants (III, IV) were optically measured to be non-plasmonic responsive between 2 – 8 μm. This fact indicates that the implanted Ga exerts low influence on the infrared radiation transmission. As such, it is determined to be non-mandatory in attempting to remove Ga post FIB milling.

Essentially, a thick metal layer is required for effective light pipe focusing in the mid infrared regime. In which, strict adherence to the critical angle rule would require the deposition of a very thick gold layer. To circumvent the limitation, instead of forming a fine taper output terminal, the structure is allowed to taper non-adiabatically with θ > θc. In this instance, the taper angle of interest has to be achievable by a typical etching process. With a half cone angle of about 10° and 20° as demonstrated by the EBL and FIB samples, the
non-adiabatic taper results in loss as evidenced by the decrease in measured transmission intensity.

However, the local field is then enhanced by the sharp corner at the terminal end of the conical hole mitigating further transmission loss. Comparing the simulated outcome between cylindrical and conical hole arrays, the latter suffers lower transmission loss (Figure 3A, Inset II). In particular, the lower loss in transmission intensity is obvious with an increase in \( r_2 \) radius. Despite its ability to preserve transmission intensity, further increase in taper angle would contribute to a decrease in the overall transmission intensity.

In order to increase the EOT effect, an overall evaluation of all participating variables have to be collectively considered. Importantly, the optimal thickness of the gold layer to support the required taper angle has to be studied. In addition, this work informs the need to optimize etching/milling process to achieve repeatable sidewall profile. This is to ensure that sensors from different manufacturing batches can be fabricated with similar performance as specified.

Previously, the differences between EBL and FIB fabricated holes have been reported by Michal et al [48]. While

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**Fig. 7.** A) Transmission characteristics of FIB samples when measuring \( \text{CO}_2 \) gas flow at different flow rates. B) Transmission and \( \text{CO}_2 \) flow rate relationship with. C) Performance comparison table showing the FWHM (FOM) of the proposed sensor against that of recent state-of-the-art mid infrared plasmonic filters with references obtained from [39]–[45]. PE–polyethylene, PVC–polyvinyl chloride, PEI–polyethylene imine.
the authors heralded the merits of FIB, rounded hole top was showcased to negatively affect the EOT performance. In contrast, as depicted in Figure 8B, greater light confinement occurs at the terminal exit end. Thus, this work demonstrates the case of rounded conical hole top exerts minimal impact on the overall field enhancement. Furthermore, cross sectional TEM analysis in Figure 5 confirms that the diameter of the terminal exit end for the FIB milled hole is comparable to that of EBL fabricated nanohole.

IV. FUTURE OUTLOOK

FIB, a physical etching tool, is showcased for the first time to form conically shaped etch holes. While FIB is an excellent tool for research purpose, it cannot be justified for mass production use. Fortunately, the lesson learnt from FIB could be readily applied to the use of dry etching techniques. Reactive ion etching is a promising and reliable way to mass produce nanohole array devices. As shown previously, dry etching parameters can be fine-tuned for precise etching [49], [50]. Being a versatile tool, a variety of plasmonic active materials such as gold, silver, copper, aluminum, etc can be etched [51]. Different gases, such as fluorine-based compounds, have been used to etch a variety of materials. The probable residual fluorine-containing coating from the dry etching process could offer a low sticking coefficient surface. In the absence of molecule accumulation on the passivated surface, the sensor is capable of performing transient molecule flow monitoring. Furthermore, the highly precise etching profile can be accomplished by dry etching. Depending on the requirement, the nanohole array can be designed and fabricated accordingly. Therefore, EOT sensors could be manufactured with commercially available etchers in the near future.

V. CONCLUSION

In this work, conical nanohole array fabrication techniques are showcased. It has been demonstrated that EBL and subsequent liftoff, and FIB processes result in 71° and 50.6° sloping metal sidewalls, respectively. The conical hole in Au gold amplifies the localized field enhancement at the acute corners resulting in the enhancement of FHWM and transmission intensity. Also, an inverted shape conical hole, where \( r_1 > r_2 \), yields a better EOT enhancement than when \( r_1 < r_2 \). Comparing \( r_1 = r_2 \) and \( r_1 = 0.625, r_2 = 0.55 \) nanohole arrays, the field enhancement of the latter is approximately 40 times that of the former. The FWHM is enhanced by 14% from 800 nm to 700 nm at 4.2 \( \mu \text{m} \) for EBL+liftoff and FIB processed nanohole array, respectively. Moreover, it has been verified that the presence of Ga on the FIB sample surface, does not contribute to a decline in the transmission profile. A CO₂ gas flow sensor is demonstrated with the highest sensitivity for CO₂ gas flow between 1–4 sccm. Furthermore, the demonstrated FIB etching methodology can be extended to reactive ion dry etching techniques where the taper angle can be precisely controlled. This case, large area nanohole array fabrication for mass production can be realized. Finally, taper angle control can be used as a new parameter to regulate the performance of EOT devices.

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