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Effect of Nanohole Spacing on the Self-Imaging Phenomenon Created by the Three-Dimensional Propagation of Light through Periodic Nanohole Arrays

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Abstract

We present a detailed study of the inter-nanohole distance that governs the self-imaging phenomenon created by the three-dimensional propagation of light through periodic nanohole arrays on plasmonic substrates. We used scanning near-field optical microscopy (SNOM) to map the light intensity distributions at various heights above 10×10 nanohole arrays of varying pitch sizes on silver films. Our results suggest the inter-hole spacing has to be greater than the wavelength of the incident light to create the self-imaging phenomenon. We also present Finite-Difference Time-Domain (FDTD) calculations which show qualitative corroboration of our experimental results. Both our experimental and FDTD results show that the self-imaging phenomenon is more pronounced for structures with larger pitch sizes. We believe this self-imaging phenomenon is related to the Talbot imaging effect that has also been modified by a plasmonic component and can potentially be used to provide the basis for a new class of optical microscopes.

Keywords

Plasmonics; Subwavelength structures; Nanostructures; Near-field microscopy; Surface plasmons; Talbot and self-imaging effects

1. Introduction

Research over the past two decades has unraveled the immense potential of the field of plasmonics and surface plasmon polariton (SPP)–based metallic nanostructures because of their promise for revolutionizing applications in a wide variety of fields such as imaging, data storage, sensing, and nanolithography.^{1–11} SPPs are electromagnetic waves that can be excited by incident light waves and are confined in the interface between materials with dielectric constants of opposite sign, a situation that commonly occurs in noble metal films in contact with a dielectric medium (such air, water, glass, quartz, etc.). At optical

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frequencies, the SPP intensity decays exponentially within less than 200 nm from the interface in the direction perpendicular to the interface, whereas SPPs can propagate along the interface.^{12–13} In the field of optical imaging, the development of a plasmonics-based microscope would have a profound impact on the biological sciences. At present there is a gap in the ability to measure distance and dimensions in the range of 1 – 250 nm, where important biological molecules and events occur. This is because optical resolution is limited to about one-half the incident wavelength ($\lambda/2$), which is a major limitation in biomedical imaging. Distances up to about 10 nm can be measured using fluorescence resonance energy transfer (FRET), but FRET does not occur at longer distances. Other methods such as Electron Microscopy (EM) cannot be used with living cells and Atomic Force Microscopy (AFM) is limited to the surface of cells and can cause deformation of the features of interest.

The optical properties of subwavelength apertures in metallic films have been the focus of much research effort since the extraordinary optical transmission (EOT) phenomenon was first reported by Ebbesen and co-workers in 1998^{14–15}. EOT is an optical phenomenon whereby a periodic array of subwavelength sized holes in an opaque metal film transmits more light than expected for nanoholes in an opaque film based on classical optics. Hence in the EOT phenomenon the holey metal film is not a mere screen that blocks the light but rather an active participant in the transmission process, whereby the collective response of the surface electromagnetic modes in the metallic film boosts the transmission. In this paper, we have performed the three-dimensional (3D) imaging of the light transmitted through periodic nanoapertures using a scanning probe to perform optical sectioning microscopy.¹⁶

Our concept for a subwavelength microscope based on plasmonic effects is shown schematically in Figure 1. This laboratory presented one of the earliest reports that illumination (green arrows) of a subwavelength sized nanohole array with monochromatic plane waves creates a self-image above the metal with the field confined to sub-diffraction-limited wavelength dimensions.¹⁶ This image is repeated at various distances above the metal. The volume of each individual spot in an imaging plane may be used to excite subwavelength volumes with lateral spot sizes on the order of 200 nm or less in a biological sample such as a cell resulting in longer wavelength fluorescence (red ovals). The individual volumes can be spaced by a micron or more so that the emission intensities can be measured for each separate volume with an imaging or single point detector. Hence the image with sub-diffraction-limited resolution can be obtained without the need for AFM or Scanning Near-field Optical Microscopy (SNOM) and the complete image of the cell can be obtained by scanning the nanohole array in the x – y plane. A 3D image can be obtained by scanning the distance of the z direction. The extent of scanning need not be large because the excited volumes are over 1 micron apart. If the spatial resolution can be brought down to 100 nm with an illuminating wavelength of $\lambda_{\text{exc}} = 514.5$ nm, then 10 images would be needed in each of the x and y dimensions, for a total of 100 images to complete one image plane. These devices would be much simpler than other approaches to subwavelength microscopy such as the Stimulated Emission Depletion (STED) method which is a resource heavy approach to break the $\lambda/2$ limit.¹⁷ Another impact of such a plasmonics-based microscopy technique would be the development of a novel class of microscopes that will not require complex and expensive glass objectives.

This study extends the work described in our previous paper where we first reported the self-imaging phenomenon created by the 3D propagation of light through periodic nanohole arrays on plasmonic substrates over distances of tens of microns above the plasmonic nanohole array.¹⁶ Here we present a detailed study dealing with their critical parameters, namely the spacing between adjacent nanoholes that govern this self-imaging phenomenon. We used SNOM to image the intensity profile of the beam transmitted through nanohole

arrays of various pitch sizes at discrete locations above the array when it is illuminated by a polarized monochromatic plane wave. We found that the period of the nanohole array plays a critical role in the generation of this phenomenon, namely the pitch size must be greater than the incident wavelength. Furthermore, increasing pitch sizes leads to formation of complex and interesting fractional image planes where the size of each spot is significantly lower than that at the surface and which lies well into the subwavelength regime. Hence this can potentially increase the opportunity to use even finer spots for super-resolution imaging applications. We also present Finite-Difference Time-Domain (FDTD) calculations which show qualitative corroboration of our experimental results regarding the formation of discrete self-imaging planes. Our FDTD results confirm that the self-imaging phenomenon is more pronounced for structures with larger pitch sizes. We believe this self-imaging phenomenon is related to the Talbot imaging effect that has also been modified by a plasmonic component and can potentially be used to excite subwavelength sized volumes in cells and thus provide the basis for a new class of optical microscopes.^{16, 18–28}

We believe the phenomenon we observed can have numerous applications to physical chemistry. The localized fields provided by nanohole array can have biophysical and/or diagnostics applications. For instance it is known that fluorescence correlation spectroscopy (FCS) can only be used with highly dilute (nanomolar) solutions, which are required to have a small number of molecules in a diffraction-limited volume, as a result FCS is often unsuitable for drug discovery applications where drug-receptor binding constants are often in the micromolar range. These weaker binding constants can become accessible if the electric fields are confined to smaller volumes to the metallic structures. The possibility of an array of such spots would allow measurements to be preferred in many cell locations in a single measurement. It seems likely that there will be many more applications beyond those mentioned above. For example, there may be applications in photolithography, data storage or clinical testing. It is remarkable that previously opaque metal films now provide opportunities for a new class of optical components based on metallic nanostructures.

It is informative to consider the potential usefulness of our observation for biological and medical research. Our results demonstrate the possibility of created highly confined light fields using only a simple metal film. Depending on the geometry of the film as spacing of the nanoholes there can be a potentially unlimited number of spots at varying distances from the metal surface. This phenomenon could be used for cell imaging by moving the metal film on sample and collecting multiple images with an array detectors followed by a single reconstruction. Even at the current spatial resolution one can imagine a new class of microscope based on such metallic optical components rather than bulky glass optics. Since the plasmon wavelengths are smaller than the free-space wavelength such metallic film could be used for sub-wavelength resolution in optical microscopy.

In more general terms plasmonic effects have become part of physical chemistry. Surface enhanced Raman spectroscopy (SERS) is mostly a plasmonic effect used in chemical research as well as for analytical applications. Surface plasmon resonance (SPR) is a plasmonic effect which reveals chemical phenomena occurring on surfaces. Changes in plasmonic resonances are used to detect proximity between metal colloids. All these effects occur in the near-field close to the metal surfaces. Our paper shows how plasmons can be used to manipulate light in the far-field and thus extend plasmonics to three dimensions.

2. Materials and Methods

2.1 Nanohole Array Fabrication and Characterization

Focused Ion Beam (FIB) milling (FEI Quanta 200 3D) was used to produce the nanohole arrays. The 10×10 arrays had a nanohole diameter of $d = 200$ nm, various pitch sizes

(center-center) of $p = 300$ nm, 500 nm, 1 μm , 2 μm and 3 μm . The arrays were written into a $t = 200$ nm thick silver film supported on glass substrates with a 30 kV, 30 pA ion beam. The nanostructured silver samples were then mounted on an aluminum stub with conductive tape, and imaged in an Hitachi SU-70 scanning electron microscope (SEM) directly, i.e., without any further treatment of the sample. Samples were surveyed at low magnifications to see the general features and the homogeneity. Representative areas were then selected for higher magnification investigation.

2.2 SNOM Imaging

The transmitted light fields were imaged using a WiTEC α -300S Scanning Near-Field Optical Microscope (SNOM) in the Collection Mode" as shown in Figure 2. The array was illuminated with a $\lambda_{\text{exc}} = 514.5$ nm Ar⁺ ion laser light (Melles Griot, Carlsbad CA). The SNOM is used to obtain the transmitted light field distribution above the array in a manner representative of optical sectioning microscopy where the SNOM tip was used to image the field intensity (E^2) on the distal side of the film by raster scanning (x - y) the sample, with the SNOM tip and objective held in a fixed position. The light collected by the SNOM tip was focused onto an Avalanche Photo-Diode (APD) detector. SNOM images were obtained with the SNOM tip located at the surface of and at various heights (z) above the 10×10 nanohole arrays.

2.3 FDTD Computational Details

Three-dimensional FDTD simulations are performed using the *FDTD Solutions* package from Lumerical Solutions, Inc. (Vancouver, Canada)^{29–32}. The FDTD code was implemented in the parallel option on a Dell Workstation. The perfectly matched layer (PML) boundary condition was applied at the simulation boundaries. In addition, symmetry and anti-symmetry boundary conditions were applied for the purpose of saving memory and for high-speed computation. After testing for convergence, we employed a grid size of 4 nm for all our calculations. The typical simulated durations of our simulations were 400 fs. The FDTD program implements a realistic frequency-dependent, lossy dielectric model for silver^{33–34}. The dimensions of the silver nanostructure modeled were as follows: array dimensions = 10×10 ; nanohole diameter $d = 200$ nm; thickness of the silver film $t = 200$ nm; nanohole arrays pitch $p = 300$ nm, 500 nm, 1 μm , 2 μm , and 3 μm respectively (center-center). The nanohole arrays were supported by a glass substrate and the entire system was suspended in air/vacuum and the incident laser beam was modeled as a plane wave of wavelength $\lambda_{\text{exc}} = 514$ nm, polarized along the x -axis, and travelling along the z -axis interacting with the plasmonic silver nanostructures from the bottom through the glass side (see Fig. 2). Field monitors were placed at discrete z positions above the surface of the nanohole arrays to detect both the intensity and spatial profile of the transmitted beam. Additional post-processing of the FDTD data was performed using *MATLAB* (version 7.0) from Mathworks (Natick, MA).

3. Results and Discussion

Figure 2a shows the SEM of a 10×10 nanohole array used in this study. The average hole diameter is $d = 200$ nm with $p = 1$ μm (1000 nm) center-center spacing. The SEM image proves that the samples we used in our imaging studies were indeed well fabricated and ordered nanohole arrays. There was an approximately 20° tilt in the imaging stage with regards to the detector and hence the square arrays have a rectangular appearance. In Figure 2b we present a schematic illustration of the experimental paradigm. The SNOM setup was used for the precise high-resolution optical mapping of the light fields generated by the transmission of the incident irradiation through the different nanohole arrays. The incident laser impinges the nanohole arrays from the bottom side through the coverslip glass support

layer. It then interacts with the plasmonic nanostructures and eventually passes through the array. This transmitted beam is then imaged at the surface of the array by the “contact mode” of the SNOM whereby the SNOM tip is in contact with the surface of the array during the $z = 0$ imaging position, and at varying distances above the array by varying the height of the SNOM tip (whereby the SNOM tip is no longer in physical contact with the array).

Figure 3a shows the topography of a 10×10 nanohole array with nanohole diameter $d = 200$ nm and pitch $p = 1 \mu\text{m}$ obtained by a SNOM tip. Although the curvature of the SNOM tip is significantly larger in size than a conventional AFM tip, Figure 3a still gives us an image that clearly shows the individual nanoholes on the silver substrate. This gives us confidence that the imaging process especially at the $z = 0$ position (where the tip is in contact with the surface of the array) did not actually cause any physical damage to the nanostructures. Figure 3b shows the SNOM field image of the nanohole array with the SNOM tip in contact with the surface of the array ($z = 0$ position). Both the field and topography image were collected simultaneously for the tip located at each discrete z position. Figure 3b shows the transmitted light coming out of each individual nanohole, but we also observe interesting “interference-like” patterns in the solid silver coated region in between the nanoholes. We believe these are SPP interference standing waves between the holes^{12–13, 15}. Interestingly, on careful observation we can notice the presence of the SPPs emanating from the edge of the last rows and columns of the array. This could be an indication for the existence of travelling plasmons being launched from the nanohole arrays. This is not too surprising as the existence of such travelling plasmons waves have been well documented^{12–13, 15}.

Figures 4(a–i) shows the SNOM field images of a 10×10 nanohole array with hole diameter $d = 200$ nm and pitch $p = 1 \mu\text{m}$ at various distances (z) above the surface of the array: (a) $z = 0$ nm – at surface; (b) $z = 1 \mu\text{m}$; (c) $z = 2 \mu\text{m}$; (d) $z = 3 \mu\text{m}$; (e) $z = 4 \mu\text{m}$; (f) $z = 5 \mu\text{m}$; (g) $z = 7 \mu\text{m}$; (h) $z = 8 \mu\text{m}$; (i) $z = 10 \mu\text{m}$. The blue size bars in all the images represents a length of $4 \mu\text{m}$. Note - images (a–f) were color scale matched to the color-bar located on their right, and images (g–i) were color scale matched to the corresponding color-bar located on their right. First, we note that the intensity does not decay sharply as we move further into z , as one may initially expect, but actually propagates along the optical axis well into the far-field. This is in agreement with our previous finding that we had not originally anticipated.¹⁶ We believe the formation of the multiple imaging planes is similar in nature to a well-known phenomenon in classical optics called the Talbot self-imaging effect which was discovered by Talbot in 1836 and later rediscovered and explained by Lord Rayleigh^{18–23}. The Talbot self-imaging effect is generally observed with periodic structures illuminated with coherent light^{18–24}. An image of the structure appears at a defined distance on the distal side of the array. It is believed that the situation is more complicated for the case of metallic nanostructures where plasmons are induced by the illumination^{16, 21–22}. For the case of a periodic structure on an opaque dielectric substrate, the Talbot phenomenon predicts that multiple images of the structure will appear at the so-called Talbot distances as expressed by^{21–22}:

$$Z_T(n) = \frac{n^2 p^2}{\lambda} \quad (n=1, 2, 3, \dots) \quad (1)$$

where n is an integer, p is the periodicity of the structure, and λ is the illumination wavelength. It is also known that a fractional Talbot effect occurs at distances of^{21–22}:

$$z = \left(\frac{n}{m}\right) Z_T \quad (n=1, 2, 3, \dots; m=1, 2, 3, \dots) \quad (2)$$

where n and m are integers. At these distances, the images are more complex than at the surface of the illuminated array, but still have a periodic structure determined by the array. For the nanohole array in Figure 4, $p = 1000$ nm, and the first and second theoretical Talbot image planes are expected to occur at $Z_T(1) = 3.90$ μm and $Z_T(2) = 7.8$ μm respectively with $\lambda_{\text{exc}} = 514.5$ nm illumination. A remarkable feature of our SNOM measurements reveal that the nanohole array shows a clear self-imaging effect at multiple z planes above the array (Figures 4b–i), which we believe is related to the Talbot plane albeit modified by a plasmonic effect^{16, 21–22}. Another interesting feature of the light intensity distribution at the different z -planes is that they appear to have a light confinement effect. The transmitted light does not diffract and diverge uncontrollably, but remains well-confined to within the array boundary of the 100 individual nanoholes. Hence we can consider a certain degree of beaming effect. Beaming of light has been predicted and observed for other structures such as a single nanohole that is surrounded by concentric rings and nanoslits flanked by periodic nanostructures^{35–39}. It is the interference of surface plasmon modes with the transmitted light that leads to the beaming phenomenon^{35–39}. We clearly see the appearance of a strong self-imaging plane at $z = 4$ μm where the nanoholes are easily resolvable and are distinct. Hence there is a striking correlation of the appearance of a strong self-imaging plane with that predicted by the Talbot plane calculations at $Z_T(1) = 3.80$ μm . Additionally, the self-imaging aspect of the silver nanohole array also displays other revival planes such as at $z = 1, 2$, and 5 μm . We believe these revival planes are related to fractional Talbot planes and appear much more pronounced for pitch sizes larger than 1 μm ($p > 1$ μm) as will be discussed in greater detail later. At values of $z > 6$ μm , the intensity of the transmitted radiation decreased and there is no appearance of a clear second image revival plane as predicted by traditional Talbot theory. Hence it appears that the transmitted field is modulated in a striking manner along the z -axis.⁴⁰ The intensity profiles demonstrate that these nanohole arrays can cause the propagating light to constructively and destructively interfere creating “dark” and “light” zones that can extend to large distances along the propagation axis.

Figures 5a–b shows the direct comparison of the SNOM field image obtained from the nanohole array of Figure 4 at $z = 4$ μm with that generated by numerical FDTD calculations. The FDTD images plots the total field intensity $|E^2| = |E_x^2| + |E_y^2| + |E_z^2|$ in the x – y plane of the transmitted beam at different z locations above the nanohole array. Of the various z locations simulated, the image generated at $z = 5$ μm bears closest resemblance to the experimental data at the first Talbot plane ($z = 4$ μm). In both images, we can clearly see transmitted light passing through the 100 nanoholes of the 10×10 array, although there is a significant difference in the intensity of the transmitted beam passing through each of the nanoholes. It is interesting to note that the light passing through the central part of the arrays appears to be significantly more intense than that passing through the peripheral region. Imperfections in the nanofabrication of the peripheral nanoholes can be ruled out as a cause for this discrepancy as the phenomenon also occurs in the numerical FDTD simulations where each nanohole in the array has identical dimensions. We believe this might be due to a plasmonic effect that might involve some sort of constructive interference of the plasmon waves from the peripheral nanoholes that concentrates energy in the central region of the array. Alternatively, the holes on the periphery have a decreased potential to interact with neighboring holes and thus contribute less to the self-image.⁴¹ The qualitative agreement between our experiments and numerical simulations gives us confidence in our observations.

Figures 6(a–i) shows the SNOM field images of a 10×10 nanohole array with hole diameter $d = 200$ nm and pitch $p = 2$ μm at various distances (z) above the surface of the array: (a) $z = 0$ nm – at surface; (b) $z = 4$ μm ; (c) $z = 6$ μm ; (d) $z = 8$ μm ; (e) $z = 13$ μm ; (f) $z = 16$ μm ; (g) $z = 18$ μm ; (h) $z = 20$ μm ; (i) $z = 30$ μm . The blue size bars in all the images

represents a length of 8 μm . The calculated first and second Talbot planes for this array are expected to occur at $Z_T(1) = 15.56 \mu\text{m}$ and $Z_T(2) = 31.13 \mu\text{m}$ respectively. The images of Figure 6 show the light intensity distribution is very complex, and exhibits many interesting features at different z locations. The intensity profiles of the transmitted light demonstrate that these nanohole arrays can cause the propagating light to constructively and destructively interfere creating “dark and light zones” that can extend as much as tens of microns along the propagation axis. We again observe the light confinement “beaming” effect with this array as we see the transmitted light does not diverge as it moves away from the substrate. We observe the appearance of several “self-imaging” planes, but in such image planes our data shows that the periodicity of the transmitted light is either greater than the original array (10×10) and/or the pitch of the period to be smaller than the image at $z = 0$. For the most part, this is in contrast to the case of the nanohole arrays with pitch $p = 1 \mu\text{m}$ as shown in Figure 4. Images with an increase in periodicity have been shown to occur with Talbot illuminators at fractional Talbot planes as defined by Eq. (2)^{21–22}, where m is an integer that reflects the periodicity. The fractional image planes at $z = 6$ and $z = 8 \mu\text{m}$ show very unique optical features, particularly the strongly confined subwavelength dimensions of the light propagating from the nanohole array. The width of each spot in the transmitted beam at these z planes is approximately 100 nm (or less) in diameter which is clearly less than that spot size on the surface of the array as shown in the cross-sectional line scan (Figure 6, bottom panel). We also see good qualitative agreement between the observation of a strong imaging plane in our experiments at $z = 18 \mu\text{m}$ and the theoretically calculated Talbot plane (for an opaque dielectric of identical parameters) at approximately $z = 16 \mu\text{m}$, although the image-plane in our experiments is not perfectly identical to the image at the surface ($z = 0$). However the intensity of the transmitted beam enters a “dark” zone at $z > 30 \mu\text{m}$ and hence does not show any pronounced self-imaging effects at such large axial distances above the array. We have reported in the past that such “dark” zones are in many cases followed by subsequent “light” zones that can extend to very large distances along the propagation axis (hundreds of microns)¹⁶. However, the purpose of this study was to focus on the self-imaging phenomenon at distances closer to the surface of the array (within the first or second “Talbot” plane) as they would be more suitable for application in a potential plasmonic microscope of the future.

Figures 7 (a–f) compares the experimental SNOM images and results of numerical FDTD calculations of the transmitted light passing through the nanohole array with pitch $p = 2 \mu\text{m}$ at various z locations. For the SNOM measurements we present images at $z = 4, 11$, and $18 \mu\text{m}$. For the FDTD calculations, we present results for $z = 6, 8$ and $14 \mu\text{m}$. Here we see a qualitative agreement between our experimental observations and theoretical calculations in 3 pairs of cases: (i) Pair 1: $z = 4 \mu\text{m}$ (SNOM) and $z = 6 \mu\text{m}$ (FDTD); (ii) Pair 2: $z = 11 \mu\text{m}$ (SNOM) and $z = 8 \mu\text{m}$ (FDTD); (iii) Pair 3: $z = 18 \mu\text{m}$ (SNOM) and $z = 14 \mu\text{m}$ (FDTD). In Figure 7 (a & b), we see an interesting agreement between the array imaged with SNOM and that calculated via FDTD. We see clearly the light transmission through each of the individual nanoholes in the array, although in the FDTD image we observe the emergence of an extra row and column of intensity spots. In Figures 7 (c and d), we see a very complex intensity patterns in both the experimental as well as the FDTD data. We do not fully understand the medley of reasons that might be behind these complex phenomena, but we suspect it involves fractional revivals the transmitted beam experiences at fractional Talbot planes. This exotic behavior is a consequence of paraxial propagation which becomes more pronounced as the pitch sizes p becomes larger than the incident wavelength of irradiation λ_{exc} ($p \gg \lambda_{\text{exc}}$).^{21–22} Although this phenomenon exists for a limited number of fractional planes, it can be sufficient to render well-defined focal spots as seen in Figure 7b that can be useful for self-imaging of small features. Figures 7 (e and f) show the emergence of a strong “self-imaging” plane for both our experiments and numerical simulations. The intensity of the focal spots in the transmitted beam is high and extremely well-defined. Additionally we

also see the emergence of an array with greater number of focal spots around the 10×10 structure than that observed at the surface of the array. Figures 6, 7 and 8 (discussed below) suggests the fact that if the pitch of the ordered nanostructure is greater than λ_{exc} the transmitted radiation will not only display multiple self-imaging or fractional self-imaging planes, but also will have complex spatial distribution of the energy of the beam. As a result even in “self-imaging” planes, it cannot be expected that the spatial profile of the transmitted beam will be identical to the surface directly above the surface of the array.

Figures 8(a–i) shows the SNOM field images of a 10×10 nanohole array with hole diameter $d = 200$ nm and pitch $p = 3 \mu\text{m}$ at various distances (z) above the surface of the array: (a) $z = 0$ nm – at surface; (b) $z = 1 \mu\text{m}$; (c) $z = 5 \mu\text{m}$; (d) $z = 7 \mu\text{m}$; (e) $z = 13 \mu\text{m}$; (f) $z = 19 \mu\text{m}$; (g) $z = 33 \mu\text{m}$; (h) $z = 42 \mu\text{m}$; (i) $z = 70 \mu\text{m}$. The blue size bars in all the images represents a length of $10 \mu\text{m}$. The calculated first and second Talbot planes for this array are expected to occur at $Z_T(1) = 35 \mu\text{m}$ and $Z_T(2) = 70 \mu\text{m}$ respectively. Similar to Figure 6, Figure 8 shows the transmitted light have very interesting and complex patterns (Fig. 8 - d,e,f) where the individual spot sizes are very small and well into the sub-diffraction limited size regime. A line scan to show the cross section of the spot sizes for $z = 19 \mu\text{m}$ is shown in the bottom panel of Figure 8. Also the periodic image pattern at these z locations has pitch sizes far smaller than that of the image on the surface. In Figure 8, we do not observe any sort of quantitative agreement in the appearance of strong “self-imaging” planes with that calculated using Talbot theory. It appears that this correspondence is a function of the ratio of pitch p to incident wavelength λ_{exc} and breaks down as $p \gg \lambda_{\text{exc}}$. It would have been very interesting to use FDTD to numerically compute the spatial distribution of the transmitted beam intensity profile as multiple z distances above the nanohole array, but unfortunately this was not possible because of limitations in computer memory required to model the larger array which had its first and second Talbot planes at $Z_T(1) = 35 \mu\text{m}$ and $Z_T(2) = 70 \mu\text{m}$ respectively. It would have been interesting to compare the deviation of the experimental data from both the FDTD calculations (in terms of spatial profile of the transmitted beam intensity as well as the z -axis positions where self-imaging planes occur) and the “self-imaging” plane locations (z) as predicted by pure Talbot theory for dielectric structures.

We have both imaged as well as used FDTD to simulate the transmitted light intensity profile at various z locations for 10×10 nanohole arrays with pitch sizes of $p = 300$ and 500 nm (center-center). In both our experiments and simulations, we found that there were no self-imaging planes formed at all (data not shown). In fact the transmitted light lost any ordered shape and we could not see any individual nanohole or even array-like features. The loss of any patterned shape was more acute for the $p = 300$ nm array than the $p = 500$ nm array. We were surprised by the loss of self-imaged planes for small spacing between nanoholes. This effect has been confirmed by numerical calculations [42]. One of the reasons we believe this is the case is because for such short pitch sizes, the area between adjacent nanoholes have large percentage of glass surface. As a result there is not sufficient silver coating present to neither: (a) serve as an opaque screen to block the light not directly impinging on a nanohole; nor (b) provide a plasmonic carpet which can actively contribute to modulate and/or boost the transmission by having its surface electromagnetic modes be collectively excited by the incident laser.

We have so far discussed and characterized the nature of light travelling along the axial propagation axis once it has been transmitted through nanohole arrays. But it is interesting to note that self-imaging also occurs along the lateral surface of such nanohole arrays once impinged by a coherent monochromatic light beam. Figure 9 shows the lateral distribution of travelling plasmons that are generated from different sized nanohole arrays when excited by the 514.5 nm line of an Ar^+ laser. On the top row we have (left-right): 7×1 , 7×3 , and 7

$\times 5$ nanohole arrays with nanohole diameter $d = 200$ nm and pitch $p = 2$ μm . On the bottom row we have (left-right): 9×1 , 9×3 , 9×5 , and 9×7 nanohole arrays with nanohole diameter $d = 200$ nm and pitch $p = 2$ μm . This result demonstrates that the Talbot effect can also occur in two-dimensions, which suggest its use to manipulate plasmons on surfaces. We investigated the light fields transmitted through the nanohole arrays in a silver film supported by a glass substrate. The array is illuminated with 514.5 nm laser light, and imaging is performed with a Witec SNOM microscope in collection mode. The SNOM is used to obtain the light field distribution above the array in a manner similar to optical sectioning microscopy. To accomplish this, the SNOM tip is initially positioned at the surface, and a SNOM image is obtained. Images were obtained by keeping the SNOM in contact with the surface of the nanostructures at all times during the image acquisition. The asymmetric launching of plasmons from the nanohole array could be related to the illumination slightly tilted from the normal. Small deviations from normal could lead to this. The images clearly reveal the generation of SPPs both inside the nanohole array as well as in the solid silver region in the vicinity of the arrays. The SPPs travel large distances (tens of μm 's) away from the site of the nanohole array. This is particularly more for the larger arrays. Additionally, the nanohole arrays with 9 nanoholes in each row (bottom set of arrays) interacts with the incident laser more effectively to create stronger SPPs as judged by both the intensity of the waves as well as their distance of travel. This is not surprising as SPPs have been reported to propagate along the metal-dielectric interface over distances of tens of μm and where also launched by subwavelength sized nanoholes on thin metal films in an arc shaped array which were then focused into an intense spot with subwavelength size^{12–13}.

4. Conclusions

In this paper we investigate the role of inter-nanohole spacing on the self-imaging phenomenon created by the three dimensional propagation of light through periodic nanohole arrays on thin silver films. The “self-imaging” effect is a complex phenomenon which is influenced by multiple factors. We aim in this paper to isolate one such parameter in a systematic study i.e. the inter-nanohole distance. We used a scanning near-field optical microscope (SNOM) to map the light intensity distributions at various heights above 10×10 nanohole arrays of identical hole sizes but varying in pitch sizes (p) on silver films caused by the transmission of 514.5 nm Ar^+ laser from the distal side. Our experiments reveal that the transmitted light does not diffract away after passing through the array, but rather displays significant modulations in the image pattern formed at different distances above the nanohole array along the propagation axis. At discrete distances that are specific to the particular array investigated, it appears that the transmitted light displays self-imaging characteristics. Additionally, the propagating fields show fairly low divergence when compared to the fields on the surface of the array with individual spot sizes being diffraction limited or even in the sub-diffraction limited regime, thus suggesting that the nanohole array might induce a field confinement effect on the transmitted beam. Our results reveal that the pitch of the nanohole arrays (the inter-hole spacing) must be greater than the wavelength of excitation for the self-imaging phenomenon to occur ($p > \lambda_{\text{exc}}$). For pitch sizes equal to or less than the incident light wavelength ($p \leq \lambda_{\text{exc}}$), we do not see the appearance of the self-imaging phenomenon. Additionally, we report that increasing pitch sizes leads to formation of complex fractional image planes where the size of each spot is significantly lower than that at the surface. This opens new opportunities to use even finer spots for potential super-resolution imaging applications. We also present results of numerical FDTD simulations that give us qualitative agreement with our experimental data regarding the formation of self-imaging planes, thus giving us confidence in their authenticity. The agreements were more accurate for higher pitch sizes ($p = 2$ μm when compared to $p = 1$ μm case). Unfortunately, the FDTD calculations could not be performed for the $p = 3$ μm pitch array because of

limitations in computer memory required to model the larger array which had its first and second Talbot planes at $Z_T(1) = 35 \mu\text{m}$ and $Z_T(2) = 70 \mu\text{m}$ respectively. The results of this study build on our previous work where we introduced the self-imaging effect with only one sample geometry (4×4 array, $d = 200 \text{ nm}$, $p = 1.6 \mu\text{m}$)¹⁶. These results further corroborate our initial findings and prove that this effect is repeatable for different kinds of sample geometries. This gives us an opportunity to investigate the principles that can lay the foundations for the potential creation of a plasmonic microscope in the future. Additional parameters we intend to study closely in the future are the thickness of the metal film, size and shape (circular or oval cross-section) of the nanoholes, and periodicity of the ordered structure. An added benefit of using FDTD calculations to gain insight into the nature of light transmission through ordered plasmonic nanostructures is that they can be used in the future to guide the nanofabrication of the appropriate nanostructures that is predicted to give the ideal response for specific applications.

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Figure 1. Schematic representation of our vision for a futuristic super-resolution optical microscope based on self-imaging of plasmonic nanohole arrays. A typical configuration would be a 200 nm thick nanohole array on a thin silver film, scanning in the x-y plane, an emission filter and a CCD camera detector.

Figure 2.

(a) Scanning electron micrograph of a 10×10 nanohole array used in this study. Nanoholes were made by focused ion beam milling into a 200 nm thick silver film thermally deposited onto a $170 \mu\text{m}$ thick coverslip. The average hole diameter d is 200 nm with $p = 1 \mu\text{m}$ center-center spacing; (b) Schematic of the WiTEC α -300S SMOM setup used for optical mapping of light fields. The SNOM tip height is varied to obtain the field images at different planes above the surface of the array.

Figure 3.

(a) SNOM topography image of a 10×10 nanohole array with hole diameter $d = 200$ nm and pitch $p = 1 \mu\text{m}$ center-center spacing; (b) SNOM field image of the nanohole array with the SNOM tip in contact with the surface of the array.

1st calculated Talbot plane at $z = 3.89 \mu\text{m}$
2nd calculated Talbot plane at $z = 7.78 \mu\text{m}$

Figure 4.

SNOM field images of a 10×10 nanohole array with hole diameter $d = 200 \text{ nm}$ and pitch $p = 1 \mu\text{m}$ (center-center) at various distances (z) above the surface of the array: (a) $z = 0 \text{ nm}$ – at surface; (b) $z = 1 \mu\text{m}$; (c) $z = 2 \mu\text{m}$; (d) $z = 3 \mu\text{m}$; (e) $z = 4 \mu\text{m}$ near – **1st Talbot plane**; (f) $z = 5 \mu\text{m}$; (g) $z = 7 \mu\text{m}$; (h) $z = 8 \mu\text{m}$; (i) $z = 10 \mu\text{m}$. The blue size bars in all the images represents a length of $4 \mu\text{m}$. Note - images (a–f) were color scale matched to the colorbar located on their right, and images (g–i) were color scale matched to the corresponding colorbar located on their right.

Figure 5.

(a) SNOM field image of a 10×10 nanohole array with hole diameter $d = 200 \text{ nm}$ and pitch $p = 1 \mu\text{m}$ (center-center) at $z = 4 \mu\text{m}$ above the surface; (b) FDTD calculation of the field intensities above the surface of an identical nanohole array at a height of $z = 5 \mu\text{m}$ above the surface. The black size bars on the bottom right side of panel (a) represents a length of $4 \mu\text{m}$.

Figure 6.

SNOM field images of a 10×10 nanohole array with hole diameter $d = 200 \text{ nm}$ and pitch $p = 2 \mu\text{m}$ (center-center) at various distances (z) above the surface: (a) $z = 0 \text{ nm}$ – at surface; (b) $z = 4 \mu\text{m}$; (c) $z = 6 \mu\text{m}$; (d) $z = 8 \mu\text{m}$; (e) $z = 13 \mu\text{m}$; (f) $z = 16 \mu\text{m}$ – **1st Talbot plane**; (g) $z = 18 \mu\text{m}$; (h) $z = 20 \mu\text{m}$; (i) $z = 30 \mu\text{m}$. All the images were color scale matched to the colorbar located on the right. The blue size bars in all the images represents a length of $8 \mu\text{m}$. (Bottom Panel) A cross-sectional line scan at a distance of $8 \mu\text{m}$ from the surface.

Figure 7.

(a) SNOM field images of a 10×10 nanohole array with hole diameter $d = 200$ nm and pitch $p = 2$ μm (center-center) at various distances (z) above the surface of the array: (a) $z = 4$ μm ; (c) $z = 11$ μm ; (e) $z = 18$ μm . FDTD calculation of the field intensities at various distances (z) above the surface of an identical nanohole array at: (b) $z = 6$ μm ; (d) $z = 8$ μm ; (f) $z = 14$ μm . The black size bar on the bottom right side of panel (a) represents a length of 8 μm .

Figure 8.

SNOM field images of a 10×10 nanohole array with hole diameter $d = 200$ nm and pitch $p = 3$ μm (center-center) at various distances (z) above the surface: (a) $z = 0$ nm – at surface; (b) $z = 1$ μm ; (c) $z = 5$ μm ; (d) $z = 7$ μm ; (e) $z = 13$ μm ; (f) $z = 19$ μm ; (g) $z = 33$ μm – **1st Talbot plane**; (h) $z = 42$ μm ; (i) $z = 70$ μm – **2nd Talbot plane**. The blue size bars in all the images represents a length of 10 μm . (Bottom Panel) A cross-sectional line scan at a distance of 19 μm from the surface.

Figure 9.

SNOM field images showing the generation and propagation of Surface Plasmon Polaritons (SPPs) around nanohole arrays of varying dimensions when excited by the 514.5 nm line of an Ar^+ laser. (Top Row: left-right): 7×1 , 7×3 , and 7×5 nanohole arrays with nanohole diameter $d = 200$ nm and pitch $p = 2$ μ m. (Bottom Row: left-right): 9×1 , 9×3 , 9×5 and 9×7 nanohole arrays with nanohole diameter $d = 200$ nm and pitch $p = 2$ μ m.