Aluminum-based fishnets with complex aperture shapes

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***Abstract*— Plasmonic structures ensure electromagnetic field localization and concentration within subwavelength volumes, thus enabling numerous practical applications. Structuring at levels much below the operating wavelength can be used to obtain novel electromagnetic modes, an example being the designer plasmon structures where one utilizes ordered arrays of subwavelength holes to mimic the behavior of metals in different frequency ranges and obtain the designer (spoof) plasmons, surface waves with a resonant frequency tailorable by design in a wide range. In this paper we consider structures with aperture-based pattern that use aluminum for the negative permittivity part instead of the usual gold or silver. We experimentally investigate patterns with complex aperture shapes which ensure richer modal behavior of the structures, including a possibility to customize their frequency dispersion and to obtain tailorable deeply subwavelength electromagnetic field hotspots. To this purpose we utilize the proximity effects (field concentration in deep subwavelength gaps) and the edge effects (localization on sharp corners). We conclude that a new degree of design freedom is obtained when utilizing aluminum-based aperture arrays and that the proximity and edge effects at a deep subwavelength level can be used to enhance the effects of spoof plasmons. It is important at that to take into account the influence of native oxide of aluminum that appears in practical structures, as well as the increased absorption losses.**

***Index Terms*— Nanophotonics; Nanoplasmonics; Aluminum Plasmonics; Extraordinary Optical Transmission Arrays; Spoof Plasmons; Nonlocality**

I. INTRODUCTION

NUMEROUS structures and devices for nanophotonics including all of those in plasmonics/nanoplasmonics base their operation on surface-bound waves [1]. Maybe the best known are the surface plasmons polaritons (SPP) [2], coupled surface waves that consists of plasmons, i.e. resonant oscillations of electron plasma in a conductor containing free electrons (usually metal) and polaritons, surface-bound p-polarized electromagnetic waves. Such waves are bound to the interface between the conductive material and dielectric, are evanescent in both directions perpendicular to the surface and may be propagating or localized, in dependence on the size of the feature where they appear [3]. The main applications of plasmonic structures stem from the fact that they ensure localization and concentration of electromagnetic field to subwavelength volumes [4]. This gives the possibility to create devices operating at optical frequencies while simultaneously having dimensions comparable to electron devices, i.e. much smaller than those unconventional photonics. In other words, the use of SPP waves may ensure packaging densities of VLSI circuitry simultaneously with THz speeds typical for photonics [5]. Other applications of plasmonic devices are found in ultrasensitive chemical or biological sensing, waveguiding, etc. [6].

A generalization of the SPP waves has been proposed by Pendry et al [7]. If a conductive surface containing free electrons is perforated by an array of apertures with subwavelength lateral dimensions, a new kind of surface bound waves appears on it. Their properties mimic very closely those of SPP, however their wavelength, propagation, even the their resonant frequency are determined by the geometry of the subwavelength hole array. Such surface waves were named spoof plasmons or designer plasmons [8]. By choosing the properties of the array one can dramatically expand the operating wavelength range. The structures utilizing spoof plasmons may function in long wavelength infrared range, terahertz and further [9], while SPPs are typically confined to the UV and visible part of the spectrum.

The subwavelength apertures themselves exhibit an apparently paradoxical effect, that of extraordinary transmission. First observed by Ebessen et al [10], the effect consists in non-negligible and even high transmission of subwavelength holes in an optically opaque (thick metal) surface, although according to the classical theory no transmission should occur at all since all polarizations should be blocked by the holes much smaller than the operating wavelength. Its very existence is a consequence of the mentioned surface waves and their coupling with propagating modes, mediated by the apertures that act as diffractive

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scattering elements. The application of spoof plasmons ensures the use of the effect of extraordinary transmission far from the visible wavelength range.

Gold and silver are used in a vast majority of cases to fabricate plasmonic structures [3]. In the last few years, however, aluminum joined the group of plasmonic materials [11], due to its lower costs, compatibility with microelectronic/microelectromechanical (MEMS) technologies and a wider spectral range compared to gold and silver, covering wavelengths from the infrared to the deep UV.

In this contribution we investigate the possibility to fabricate structures able to support spoof plasmons using aluminum for the plasmonic parts. The structures utilize complex shapes of subwavelength apertures in order to make use of the electromagnetic field hotspots and thus to obtain a richer spectral response. Experimentally, we use radiofrequent sputtering to deposit aluminum to our silicon wafers with thermal silicon dioxide layer and laser writer-based photolithography to directly draw the complex aperture shapes in photoresist. The structures are designed for the mid infrared wavelength range. We utilize Fourier-transform infrared microscope to measure the spectral characteristics of the spoof plasmon samples.

II. Aluminum-Based Plasmonics

For a number of years the materials of choice for plasmonics have been gold and silver [3]. They are often described as good metals, meaning that their free electron concentration is high while the absorption losses are lower than in other metals. In recent years, however, researchers started introducing new, alternative plasmonic materials like transparent conductive oxides, intermetallics, heavily doped semiconductors, etc. [12, 13].

One of the alternative plasmonic materials currently attracting large interest of the research community is aluminum [11, 14]. It was actually briefly used in 1959, at the very beginnings of the plasmonics era, to be immediately replaced by gold and silver. It re-emerged only several years ago.

Compared to silver or gold, aluminum reaches negative dielectric permittivity over a wider frequency range. One of the important points of interest for its use as plasmonic material is nonlinear optics [11], because of the ability of plasmonic structures to localize and concentrate optical fields for several orders of magnitude and thus strongly enhance nonlinear effects.

One of the problems with aluminum is its oxidation in air. A native oxide layer is built on Al surface immediately after it has been exposed to atmospheric oxygen (of the order of several minutes), with a thickness of about 3 nm. To avoid bulk oxidation, a protective dielectric coating is often deposited over aluminum surface.

Large discrepancies were observed between theoretical and experimental data on aluminum plasmonics. Knight et al [14] successfully explained this by the presence of oxide in bulk. A larger amount of oxide causes a redshift of the spectral dependence and decreases electromagnetic scattering. Structures with more than 50% of oxide do not exhibit any plasmonic resonance at all.

Besides the appearance of oxide on the surface and in bulk impairing its plasmonic performance, another important disadvantage of aluminum are the mentioned increased absorption losses. They are much larger than in silver or gold and impair the performance of aluminum-based plasmonics.

Figure 1 shows the spectral dispersion of the relative dielectric permittivity of aluminum. The peaks in both real and imaginary part of permittivity around 800 nm are a consequence of interband transitions. Fig. 2 presents the reflection coefficient of aluminum in ultraviolet, visible and infrared range, showing that the imaginary component of the permittivity increases with the wavelength.

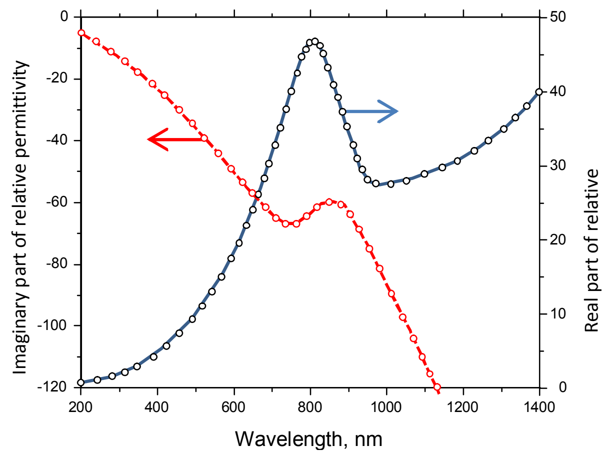

Fig. 1. Real and imaginary part of relative dielectric permittivity of aluminum in UV, visible and near IR part of the spectrum.

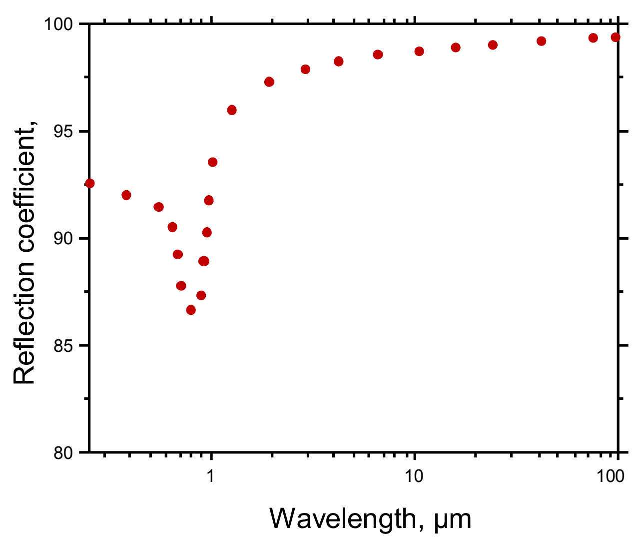

Fig. 2. Reflection coefficient of aluminum in ultraviolet, visible and infrared range.

III. Spoof Plasmon Structures

Perforated metal surfaces intrinsically ensure high field concentration within the apertures, even several orders of magnitude compared to the free space field [2]. However, in order to further increase local fields in apertures, we considered hole shapes with deep subwavelength features that promote the appearance of additional field hotspots [15]. Among such geometries are sharp edges and narrow gaps. Because of this we analyzed two types of complex-shape unit cells. Both were fabricated by twice superposing a basic form, in this case a simple square, and making a hole in aluminum layer shaped as the superposed form. If the superposition was done with a partial overlap, we obtained the structures with an increased number of sharp corners (6 instead of 4), as shown in Fig. 3 left. If no overlap occurred, but the basic squares were very close to each other, we obtained a narrow gap between them, as shown in Fig. 3 right.

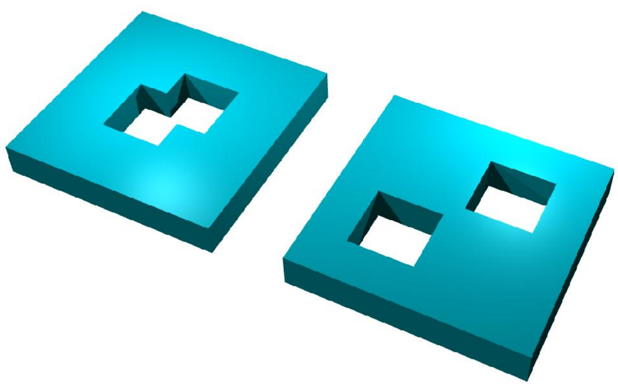

Fig. 3. Schematic presentation of the complex unit cells of aluminum-based subwavelength aperture arrays. Left: overlapping squares. Right: separate squares with a narrow gap between.

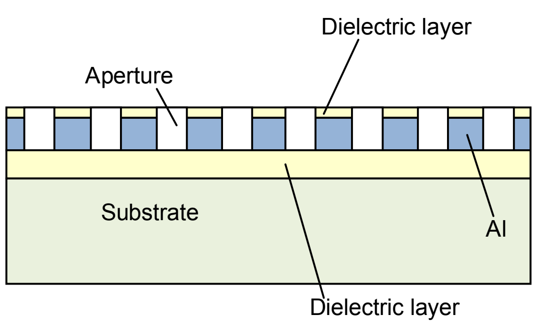

Fig. 4. Cross-sectional profile of aluminum-based single metallic layer (single fishnet, EOT).

Regarding the profile of the structures, we chose to consider EOT with single opaque metal layer – single fishnet, i.e. the conventional EOT, Fig. 4a. The structures were deposited on a semiconductor substrate with a dielectric layer on top of it and both were capped with protective oxide layer.

IV. Experimental

The layout of spoof plasmon structures that we used to produce our samples is shown in Fig. 5. It can be seen that the 2D lattice has been rectangular. One pattern utilized overlapping squares with 8 μm sides, with a shift of 4 μm along the horizontal and the vertical direction. The other pattern had unit cells with two neighboring squares, the distance along vertical and horizontal direction between the two closest edges being 2 μm each. The side of the square unit cell in both cases was 24 μm.

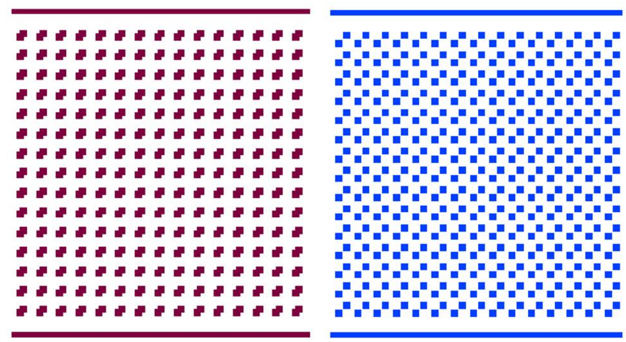

Fig. 5. Pattern layout of subwavelength hole structures. Left: unit cells with overlapping squares. Overlap is 4 μm along x and y direction. Right: unit cell consisting of two squares with neighboring edges. The shift between two squares in a single unit cell is 2 μm both along x and y direction. In both cases the basic square side is 8 μm, and the square unit cell side is 24 μm.

As for the profile of our structures, we utilized a single-layer aluminum film. The thickness of the aluminum layer was 500 nm.

We started our experiments from single-side polished n-type single crystalline silicon wafers with <100> orientation, 2-5 Ωcm resistivity. A 600 nm thick thermal oxide layer has been grown on it. Radiofrequent sputtering of aluminum in inert atmosphere of argon was further used to form a 500 nm thick aluminum layer over the thermal oxide. The exposure of the structure to atmospheric oxygen immediately results in the appearance of a surface layer of native oxide, which has to be taken into account when calculating the properties of the structures. A layer of AZ1505 photoresist 0.5 μm thick has been further spin-coated onto the aluminum surface. The photolithography of the pattern shown in Fig. 5 has been done using LaserWriter LW 405. Finally the aluminum layer has been completely removed in the position of complex-shaped holes by isotropic chemical etching.

Microscopic photographs showing the obtained geometry for the neighboring squares are shown in Fig. 6, and for the overlapping squares in Fig. 7. Two magnifications are given in each of the figures. It can be seen that the sharp corners are somewhat smoothed, which is a consequence of the implemented photolithographic procedure, i.e. both the influence of the available resolution of the LaserWriter and the etching process.

Spectral transmission of the obtained samples was measured utilizing a Fourier transform infrared microscope iN10 Thermo Scientific Nicolet with a mercury cadmium telluride detector cooled by liquid nitrogen, the spectral range being between 2.5 µm and 15 µm. Silicon-silica pair has been used as the substrate, in spite of both materials having large absorption losses at longer wavelengths, because of their properties being compatible with the microelectronic and MEMS technology. The problems of the excessive transmission loss were dealt with by using submicrometer values of thickness for both, thus decreasing the optical path through the absorptive media.

The transmission was determined by first measuring the spectral characteristics of a silicon wafer with a thermal oxide overlayer, but without aluminum. This spectrum was then utilized as the background for the measurement of the holey structures.

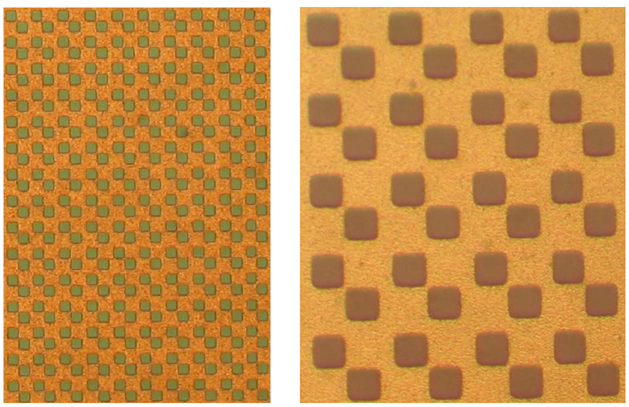

Fig. 6. Microphotographs of samples with hole pairs with narrow gaps between them; dimensions as in Fig. 5 right.

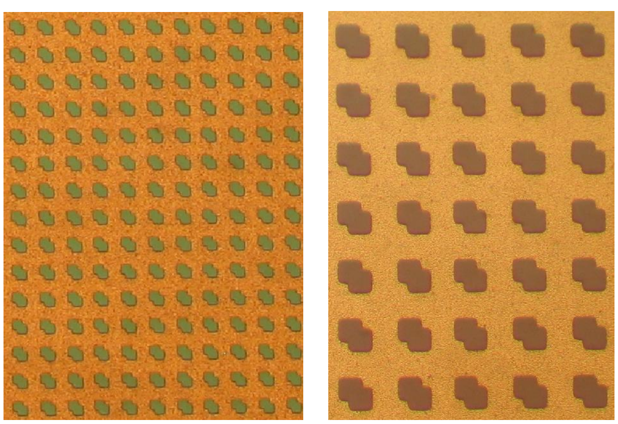

Fig. 7. Microphotographs of samples with overlapping hole pairs; dimensions as in Fig. 5 left.

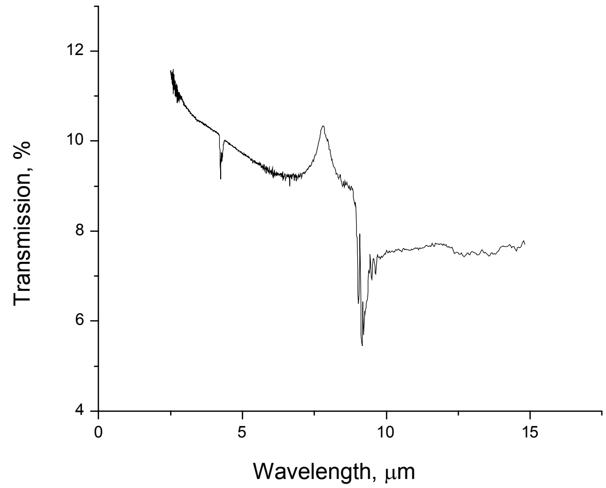

Fig. 8. Measured transmission spectrum for overlapping square holes; dimensions as in Fig. 5 left.

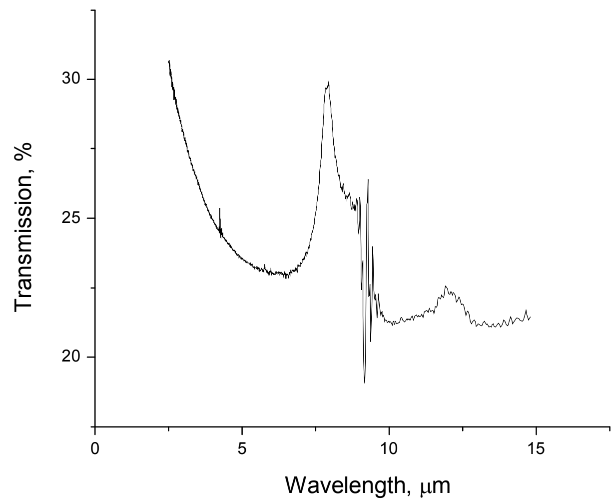

Fig. 9. Measured transmission spectrum for neighboring square holes; dimensions as in Fig. 5 right.

Figure 8 shows the spectral dependence of the transmission for the case of overlapping square holes, while the spectral transmission of neighboring holes is shown in Fig. 9. It can be seen that the transmission through the neighboring hole pairs is almost three times larger than that through the overlapping squares, although in the neighboring case the hole area is only one third larger than in the overlapping case, the rest of the surfaces in both cases being covered by an opaque layer of aluminum.

The kink at about 4 µm is caused by the presence of carbon dioxide, while the instability near 9 µm is a signature of the presence of silicon dioxide as well as water vapor. A transmission peak can be seen near 8 µm, and a weaker peak is observed at 12 µm. The available cooled mercury cadmium telluride photodetector did not allow us to measure spectral transmissions at wavelengths above 15 µm.

V. CONCLUSION

We considered complex subwavelength aperture arrays in optically thick (500 nm) aluminum film. The unit cell consisted of a combination of two square apertures with 8 μm sides, with either an overlap or formation of a narrow gap. This ensured the appearance of electromagnetic field hotspots due to edge and proximity effects. The structures are designed to be able to support plasmon-like surface waves, at the same time exhibiting a richness of the obtained modes and the tailorability of the spectral dependence of the scattering parameters.

Some problems have been observed connected with the available technology used for the fabrication of the samples. This includes a relatively coarse resolution of the laser writer equipment, its minimum feature being 1 μm, as well as inaccuracies introduced by the wet etching procedure. Another problem are the used substrates which are lossy at longer wavelengths. Still, transmission maxima have been observed in the measured spectral characteristics, including the weak peak in the subwavelength range.

We conclude that a new degree of design freedom is obtained when utilizing aluminum-based aperture arrays with deep subwavelength inclusions and that the proximity and edge effects can be used to enhance the effects of spoof plasmons.

Our further works should include the fabrication of samples with smaller features, as well as the first experiments with the application of the holey structures in sensing.

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