Communication

Numerical Investigation of a High-Quality Factor Refractometric Nano-Sensor Comprising All-Dielectric Metamaterial Structures

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Abstract: This paper proposes an optical sensor based on nanoscale metamaterial structures. The design of the sensor has been explored with respect to biosensing applications through numerical modeling and analysis. The sensor comprises silica substrate and diamond nanostructures, both of which represent dielectrics. The sensing principle is based on the detection of ambient refractive index change. As the analyte properties change, the refractive index changes, as well. The refractive index change has been detected by striking electromagnetic waves onto the structure and noting the spectral response. Ultraviolet waves have been utilized for recording spectral responses and evaluating sensor performance. The sensor displays multiple sharp resonance peaks in the reflected beam. By altering the refractive index of the analyte present around the sensor, the peaks can be seen choosing different wavelengths. The resonance peaks have been investigated to observe electric and magnetic field dipoles in the sensor structure. The spectrum peaks have also been studied to understand fabrication tolerances. The sensor displays a linear response, along with a large Quality (Q) factor. The maximum value of the achieved Quality (Q) factor for the proposed sensor is 1229 while operating across the refractive index range of 1.4–1.45. The claim has been supported by comparison with contemporary works on similar platforms. A range of other sensing parameters have also been calculated and benchmarked. Metamaterial-based optical sensors can provide smaller device sizes, faster response times and label-free detection.

Keywords: diamond; metasurface; biosensor; refractive index; ultraviolet

1. Introduction

The manufacture of glass dates back to 3000 B.C.E. The traders near the Mediterranean Sea used the sand available on the Canaanite–Phoenician coast to make glass. The glass produced at that time was opaque because of trapped particles within the structure, giving rise to the need for material engineering. Artificial engineering of glass is believed to have started in the first century B.C.E. Hot kilns were used to make glass. Artisans added metal oxides and charcoal to glass to make different colors of glass. Hundreds of years later, a classic piece of evidence of novel light matter interaction was developed, which is famously known as the Lycurgus Cup. It was created during the 4th century A.D. in the Roman Empire. The cup looks red when illuminated from the back and green when illuminated from the front. Such behavior is attributed to the introduction of gold and silver nanoparticles into glass. The Lycurgus Cup probably shows the commencement of the field of plasmonics via the use of metals with dielectrics [1,2].

Even after the creation of the Lycurgus Cup, the concept of negative refractive index materials was not formally explained until 1968. In that year, Professor Viktor Georgievich
Veselago proposed a theoretical model for optical materials, with negative values of electrical permittivity and magnetic permeability both [3]. Veselago’s work started to gain its due attention when plasmonics and metamaterials grew into a wondrous scientific discipline around the start of the twenty-first century.

Hence, the present-day field of metamaterials is a result of hundreds of years of research. Yet, it does not stop surprising researchers with its tremendous potential. In fact, it seems that active research on optical metamaterials has just taken an exciting new turn during the past couple of decades. More than 5000 publications have been dedicated to the study of optical and negative-index metamaterials over the years 2000 to 2011 [4–6].

In light of Veselago’s work, it can be believed that a metamaterial is basically a material having negative permittivity (\(\varepsilon\)) and permeability (\(\mu\)). In other words, a metamaterial can be called a ‘Double Negative (DNG) Material’ or ‘Left-Handed Material (LHM)’. Modern research has, however, emphasized that the scope of metamaterials is broader than LHM and DNG. In today’s era, metamaterial is used to describe an artificially engineered material with exotic properties exhibited by unit cell structures that tailor the wavefronts of electromagnetic waves at a scale smaller than the wavelengths of incident waves [7]. The general expression relating the refractive index of a metamaterial to permittivity (\(\varepsilon\)) and permeability (\(\mu\)) is given below.

\[
\frac{1}{n} = \pm \sqrt{\frac{\varepsilon}{\mu}}
\]  

The past two decades have witnessed a humongous interest in metamaterials. A particular reason is the birth of flat optics. Flat optics deals with the design of metamaterials at subwavelength scale in the form of nanometer-sized structures [8,9]. The nanostructures can be arranged in different patterns for specific applications. Patterns of such optically resonant structures on flat layers of materials are called metasurfaces. Metasurfaces can be considered to be two-dimensional counterparts of metamaterials. Parameters associated with incident light, such as phase, polarization and amplitude, can be tuned by the nanomaterial resonators present on the metasurface.

Metasurfaces have shown extraordinary potential in several applications, which include communications, automobiles, biomedical imaging and sensing. This paper focuses on the potential of metasurface-enabled biosensing, which provides economical and label-free methods in many cases. A meta-device or meta-sensor has a much smaller footprint compared to traditional equipment. The response time and sensitivity are also appreciable. Metasurface biosensors have been proposed for a variety of chemicals, including glucose, hemoglobin, ethanol and ethylene glycol. Biomarkers indicating the presence of certain diseases have also been investigated through metamaterial biosensing platforms [10–18]. Reference [14] has presented a thorough review on plasmonic nanostructures for sensing applications. The review has highlighted the significance of point-of-care (POC) diagnostic capabilities. It has also listed a number of techniques, materials, design shapes and particles that have been targeted under the domain of plasmonic sensing. It is worth noting that gold and silver have appeared to be the popular choice in most of the works cited by the paper. A wide variety of biomarkers for different diseases have been discussed, including cancer, COVID-19, Hepatitis, etc. This paper has also provided key information on signal read-out methods, which is an important perspective for the industrial applicability. A variety of other sensing tasks have been achieved by plasmonic metasurfaces. Patel et al. have presented a thorough perspective on the role of metasurfaces in the detection of COVID-19. Graphene has recently shown widespread use in metasurface biosensors. Many groups have targeted graphene while designing biosensors [19–23].

It is worth noting that the Surface Plasmon Resonance has been a more famous candidate for biosensing. Recently, this observation has been brought to light, and some researchers have started investigating the role of dielectric resonances in biosensing platforms. A paper [24] proposing dielectric nanostructures for a sensing application has acknowledged the fact that Surface Plasmon Resonance in metallic nanostructures provides the capability of achieving higher sensitivity. The literature in the preceding section also validates this acknowledgement. However, the paper also mentions that the Joule heating
effect is inevitable in metals. This leads to higher losses. Here, it is important that metallic metasurfaces with low losses have also been reported recently [25,26]. Further, the dielectric structures possess stronger resonance properties. As opposed to metallic optical antennas, dielectric antennas show Mie resonance. Dielectric biosensors offer another advantage over plasmonics, which is the compatibility with Complementary Metal Oxide Semiconductor (CMOS) processes. Despite these advantages, research on dielectric biosensing metasurfaces has not been as rampant as that on plasmonic metasurfaces [27].

Therefore, in this paper, the complete focus of design is on the use of all-dielectric metasurfaces. A novel metasurface has been presented, consisting of diamond nanodisks placed on silica substrate. Diamond metasurfaces have shown promising results through Mie resonances in the ultraviolet regime [28], but they have hardly been used for application-specific sensing problems. Here, the application being considered is the concentration sensing of aqueous glucose. The ultraviolet frequency spectrum can prove to be highly potent for sensing problems due to the high energy level. Its use becomes more relevant in the context of sub-wavelength optics since they have smaller wavelengths compared to the visible and infrared (IR) spectra. This means that a UV sensor can be more compact than visible and IR sensors. It can also have a better resolving power, while detecting very minute molecules that have dimensions of the same scale as UV wavelengths. Table 1 lists all the recent works reported on the sensing of aqueous glucose through metasurface optics. It is clear that sensing through ultraviolet frequencies is a rarely implemented concept in the realm of planar optical elements, despite the advantages mentioned earlier. It is also worth noticing that the investigation carried out in this paper yields favorable results in terms of the sensor characteristics. In this work, a thorough analysis has been presented, in terms of sensor performance, for the application being considered. The use of resonant nanostructures for sensing minute biochemicals through the ultraviolet frequencies, along with the promising sensor performance features, make this work novel. Aqueous glucose sensors find plentiful applications in the food and biomedical industries. Aqueous glucose sensing ability may also be extended for blood glucose prediction. The sensing performance depends on the change in the refractive index of aqueous glucose.

Table 1. Contemporary Literature on Meta-Optic Glucose Sensors.

<table>
<thead>
<tr>
<th>REF.</th>
<th>FREQUENCY BAND</th>
<th>MATERIALS</th>
<th>STRUCTURE</th>
<th>SPECTRAL REFERENCE</th>
<th>REFRACTIVE INDEX RANGE</th>
</tr>
</thead>
<tbody>
<tr>
<td>[29]</td>
<td>Visible</td>
<td>SiN, Si</td>
<td>Nano-rod</td>
<td>Reflection Peak</td>
<td>1.33–1.38</td>
</tr>
<tr>
<td>[30]</td>
<td>Near-Infrared</td>
<td>Si, SiO₂</td>
<td>Nano-disk</td>
<td>Transmission Peak</td>
<td>1.0–1.4</td>
</tr>
<tr>
<td>[31]</td>
<td>Terahertz</td>
<td>Au, SiO₂, graphene</td>
<td>Cuboid</td>
<td>Absorption Peak</td>
<td>1.0–2.0</td>
</tr>
<tr>
<td>[32]</td>
<td>Terahertz</td>
<td>Au, SiO₂, graphene</td>
<td>Cuboid</td>
<td>Absorption Peak</td>
<td>1.3198, 1.3594</td>
</tr>
<tr>
<td>[33]</td>
<td>Near-Infrared</td>
<td>Au, SiO₂, BK₇ glass</td>
<td>Ring-shaped nano-hole</td>
<td>Reflection Dip</td>
<td>1.32–1.5</td>
</tr>
<tr>
<td>[34]</td>
<td>Near-Infrared</td>
<td>Au</td>
<td>Split ring, nano-cylinders</td>
<td>Absorption Peak/Reflection Dip</td>
<td>1.305–1.345</td>
</tr>
<tr>
<td>[36]</td>
<td>Near-Infrared</td>
<td>Au, Ag, Cu, Al, MgF₂, SiO₂, HPDE, Al₂O₃, PMMA</td>
<td>Nano-ring, nano-disk</td>
<td>Absorption Peak</td>
<td>1.33–1.4393</td>
</tr>
<tr>
<td>[37]</td>
<td>Near-Infrared</td>
<td>Au, Ag, Cu, Al, MgF₂</td>
<td>Nanostructure with multiple layers and circular well for sensing material</td>
<td>Absorption Peak</td>
<td>1.34–1.45</td>
</tr>
</tbody>
</table>
Table 1. Cont.

<table>
<thead>
<tr>
<th>REF.</th>
<th>FREQUENCY BAND</th>
<th>MATERIALS</th>
<th>STRUCTURE</th>
<th>SPECTRAL REFERENCE</th>
<th>REFRACTIVE INDEX RANGE</th>
</tr>
</thead>
<tbody>
<tr>
<td>[38]</td>
<td>Near-Infrared</td>
<td>Ag, SiO₂</td>
<td>Nano-cylinder</td>
<td>Absorption Peak</td>
<td>1.33–1.37</td>
</tr>
<tr>
<td>[39]</td>
<td>Terahertz</td>
<td>Al</td>
<td>S Shape</td>
<td>Transmission Peak/Reflection Dip</td>
<td>1.2–2.0</td>
</tr>
<tr>
<td>[40]</td>
<td>Near-Infrared</td>
<td>Au, Si</td>
<td>Nano-bars</td>
<td>Transmission/Absorption Peaks</td>
<td>1.33–1.39</td>
</tr>
<tr>
<td>[41]</td>
<td>Near-Infrared</td>
<td>Au, MgF₂</td>
<td>Plus Shape</td>
<td>Reflection Dip/Absorption Peak</td>
<td>1.312–1.384</td>
</tr>
<tr>
<td>[42]</td>
<td>Terahertz</td>
<td>Al, SiO₂</td>
<td>Split Ring Resonator (SRR)</td>
<td>Transmission Dip/Analyte Absorption Curves</td>
<td>1.2–2.0</td>
</tr>
<tr>
<td>This Paper</td>
<td>Ultraviolet</td>
<td>Diamond, SiO₂</td>
<td>Nano-disk</td>
<td>Transmission Dip/Reflection Peak</td>
<td>1.4–1.45</td>
</tr>
</tbody>
</table>

2. Materials and Methods

The proposed biosensor design has been presented in Figure 1, along with the material dispersion properties. The dispersion model for silica [43] has been obtained from the equation given below. The wavelength unit for the formula is microns.

\[
n^2 = \frac{0.6961663\lambda^2}{\lambda^2 - 0.0684043^2} + \frac{0.4079426\lambda^2}{\lambda^2 - 0.1162414^2} + \frac{0.8974794\lambda^2}{\lambda^2 - 9.896161^2} + 1
\]  

Figure 1. (a) The metasurface unit cell with marked geometrical parameters. (b) Real and (c) Imaginary dielectric constants for the materials used in the nano-sensor. (d) Reflection peaks and (e) Corresponding Transmission dips. (f) The 3-dimensional electric field (V/m) radiation plot at a wavelength of 368.3 nm (1 of the 2 reflection resonance peaks).
The diamond’s optical parameters have been obtained from [44,45]. The dielectric constants ($\varepsilon$) of the materials depend on the frequency ($\omega$) of operation and have two components, real ($\varepsilon_1$) and imaginary ($\varepsilon_2$), as shown by Figure 1b,c. Here, it can be noted that the imaginary part of the dielectric constant, which is related to loss, goes to zero at about 220 nm for diamond.

$$\varepsilon(\omega) = \varepsilon_1(\omega) - j\varepsilon_2(\omega)$$  \hspace{1cm} (3)

Figure 1 shows that two resonance peaks in the reflection spectrum, both within the ultraviolet regime, have been achieved. By designing a multi-resonant metasurface sensor, the concentration of the analytes can be tracked through multiple peaks, providing a validation mechanism. In the figure, the radiation plot for one of the two peaks has also been provided to explain how the reflection of the electric field takes place. It can be observed that almost no electric field is transmitted at the resonance wavelength. The radiation plot for the other peak would also match closely with the one presented.

The reflection peaks correspond to the generation of electric and magnetic dipoles, as anticipated by the Mie resonance theory. The metasurface provides a strong electric dipole at a wavelength on the lower resonant wavelength, while a magnetic dipole can be observed on the higher resonant wavelength. The plane of view is x-y for Figure 2.

FIGURE 2. (a) Electric dipole (V/m) and (b) Magnetic dipole (A/m) at wavelengths of 319.78 nm (first resonance wavelength) and 368.3 nm (second resonant wavelength), respectively. The scales represent values in decibels. The views relate to the x-y plane.

The nano-sensor has been simulated through the Finite Difference Time Domain (FDTD) method. Periodic boundary conditions have been used for the simulations. The incidence of light occurs in the negative z direction. Plane wave excitation has been used, along with linear polarization. The mesh type is tetrahedral. Parametric sweeps have been employed to optimize the structure. By processing the Scattering (S) Parameters of the unit cell, the reflection and transmission plots can be obtained, as shown in Figure 1. To observe the spectral shift, simulations have then been repeated for slightly varying refractive indices of the analyte, representing different concentrations of glucose.

$$R(\omega) = |S_{11}|^2$$  \hspace{1cm} (4)

$$T(\omega) = |S_{21}|^2$$  \hspace{1cm} (5)

The following values have been used for the sensor parameters shown in Figure 1a: P = 340 nm, H = 100 nm, d = 150 nm and h = 15 nm. It is important to understand how the parameter variations of the unit cell may affect the sensor performance. For this purpose, slight variations of the metasurface parameters have been investigated. The effects of the variations on the targeted ultraviolet spectral responses have been recorded in Figure 3. The
analysis reveals that the sensor is tolerant of minor variations that may occur during the fabrication process, but large variations adversely affect the performance of the structure.

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\[
R(\chi) = |S_{11}|^2 \quad (4) \\
T(\chi) = |S_{21}|^2 \quad (5)
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Figure 3. Effects of varying geometrical parameters on resonance peaks.

3. Results and Discussion

The metasurface has been tested for sensing aqueous glucose concentration levels. The refractive index (n) of glucose solution changes with the concentration (C in g/100 mL), according to the equation below. The refractive index of water (\(n_w\)) equals 1.33 at room temperature.

\[n = n_w + 0.00143 \cdot C \quad (6)\]

Figure 4 shows how the spectra shift in the presence of the analyte. The analyte chamber has been placed on top of the metasurface. It spans across the metasurface with dimensions 170 nm \(\times\) 170 nm \(\times\) 150 nm. Here, it is important to observe that recent progress in flat optical biosensors has tremendously decreased the need for large volume sensing.

After plotting the spectral responses for different concentration levels of glucose, it is important to analyze the sensor in terms of quantifiable performance characteristics. The sensor characteristics have been calculated for both the resonant wavelengths, as shown by Table 2. Using the formulas for sensor characteristics, the maximum sensitivity displayed by the sensor is around 110 nm/RIU, where RIU stands for Refractive Index Unit.

\[
\text{Sensitivity (S)} = \frac{\Delta \lambda}{\Delta n} \quad (7) \\
\text{FOM} = \frac{S}{FWHM} \quad (8) \\
Q = \frac{\lambda_{\text{res}}}{FWHM} \quad (9) \\
\text{Detection Limit (DL)} = \frac{\Delta n}{1.5} \cdot \frac{FWHM}{\Delta \lambda} \cdot 1.69 \quad (10) \\
\text{Dynamic Range (DR)} = \frac{\lambda_{\text{res}}}{\sqrt{FWHM}} \quad (11) \\
\text{Detection Accuracy (DA)} = \frac{1}{FWHM} \quad (12)
\]

Figure 4. (a) Biosensor response to changing refractive indices. (b) Regression analysis of the two resonance peaks.
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\text{Sensitivity (} S \text{)} = \frac{\Delta \lambda}{\Delta n}
\]

(7)

\[
\text{FOM} = \frac{S}{\text{FWHM}}
\]

(8)

\[
Q = \frac{\lambda_r}{\text{FWHM}}
\]

(9)

\[
\text{Detection Limit (} DL \text{)} = \left( \frac{\Delta n}{1.5} \right) \left( \frac{\text{FWHM}}{\Delta \lambda} \right)^{1.25}
\]

(10)

\[
\text{Dynamic Range (} DR \text{)} = \frac{\lambda_r}{\sqrt{\text{FWHM}}}
\]

(11)

\[
\text{Detection Accuracy (} DA \text{)} = \frac{1}{\text{FWHM}}
\]

(12)

\[
\text{Signal to Noise Ratio (} SNR \text{)} = \frac{\Delta \lambda}{\text{FWHM}}
\]

(13)

\[
\text{Resolution (} R \text{)} = S \times DL
\]

(14)

\[
\text{Uncertainty (} \chi \text{)} = 2(\Delta \lambda)^{0.75}(\text{FWHM})^{0.25}\frac{9}{9}
\]

(15)

In the equations above, FWHM stands for Full Width at Half Maximum, and \( \lambda_r \) represents resonant wavelength. \( \Delta \lambda \) and \( \Delta n \) signify the differences in the reflection peak wavelengths and refractive indices, respectively.

Figure 4 shows the shifts in the resonance wavelengths because of variations in the ambient refractive indices. The spectral shifts have been used to perform regression analysis that yields linear relationships between the refractive index and resonant wavelengths. Larger shifts in the lower-frequency peak can be observed, providing higher sensitivity.

Table 2. Sensor Performance Parameters.

<table>
<thead>
<tr>
<th>SENSOR CHARACTERISTICS</th>
<th>PEAK 1</th>
<th>PEAK 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Full Width at Half Maximum (FWHM)</td>
<td>0.26 nm</td>
<td>0.45 nm</td>
</tr>
<tr>
<td>Sensitivity (S)</td>
<td>67 nm/RIU</td>
<td>100 nm/RIU</td>
</tr>
<tr>
<td>Figure of Merit (FOM)</td>
<td>257,692 RIU−1</td>
<td>222,222 RIU−1</td>
</tr>
<tr>
<td>Quality (Q) Factor</td>
<td>1229.269</td>
<td>769.291</td>
</tr>
<tr>
<td>Detection Limit (DL)</td>
<td>2.033 RIU</td>
<td>2.283 RIU</td>
</tr>
<tr>
<td>Dynamic Range (DR)</td>
<td>626.807 nm(^{1/2})</td>
<td>516.056 nm(^{1/2})</td>
</tr>
<tr>
<td>Detection Accuracy (DA)</td>
<td>3.846 nm(^{-1})</td>
<td>2.222 nm(^{-1})</td>
</tr>
<tr>
<td>Signal-to-Noise Ratio (SNR)</td>
<td>1.977</td>
<td>1.802</td>
</tr>
<tr>
<td>Resolution (R)</td>
<td>136.211 nm</td>
<td>228.300 nm</td>
</tr>
<tr>
<td>Uncertainty</td>
<td>0.0963 nm</td>
<td>0.156 nm</td>
</tr>
</tbody>
</table>
In Table 3, the proposed sensor performance has been benchmarked against other latest works reported in the literature. The comparison table list contains all-dielectric sensors. It can be seen that the proposed design is novel because of the resonance frequencies, as well as the commendable quality factor.

Table 3. Comparison of the Proposed Work with Other All-Dielectric Sensors.

<table>
<thead>
<tr>
<th>REF</th>
<th>OPERATING FREQUENCY</th>
<th>STRUCTURE</th>
<th>S (nm/RIU)</th>
<th>FOM (RIU$^{-1}$)</th>
<th>Q</th>
</tr>
</thead>
<tbody>
<tr>
<td>[46]</td>
<td>Mid-Infrared</td>
<td>Lucky knot</td>
<td>986</td>
<td>32.7</td>
<td>520</td>
</tr>
<tr>
<td>[47]</td>
<td>Visible</td>
<td>Rectangular posts</td>
<td>192</td>
<td>64</td>
<td>-</td>
</tr>
<tr>
<td>[48]</td>
<td>Visible</td>
<td>Periodic grating</td>
<td>82.29</td>
<td>433.1</td>
<td>3207.9</td>
</tr>
<tr>
<td>[49]</td>
<td>Long-Wave Infrared</td>
<td>Asymmetric nano-holes etched in square Si structure</td>
<td>2803</td>
<td>350</td>
<td>-</td>
</tr>
<tr>
<td>[50]</td>
<td>Mid-Infrared</td>
<td>Square nano-disk</td>
<td>1430</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>[51]</td>
<td>Near-Infrared</td>
<td>Elliptical nano-cylinders</td>
<td>400</td>
<td>3074</td>
<td>&gt;10$^4$</td>
</tr>
<tr>
<td>[52]</td>
<td>Near-Infrared</td>
<td>Rectangular bar with a ring</td>
<td>289</td>
<td>103</td>
<td>-</td>
</tr>
<tr>
<td>[53]</td>
<td>Near-Infrared</td>
<td>Nano-blocks</td>
<td>306.71</td>
<td>10.09</td>
<td>-</td>
</tr>
<tr>
<td>[54]</td>
<td>Near-Infrared</td>
<td>U-shaped cylinder</td>
<td>203</td>
<td>29</td>
<td>130</td>
</tr>
<tr>
<td>[55]</td>
<td>Near-Infrared</td>
<td>Split ring</td>
<td>452</td>
<td>56.2</td>
<td>133</td>
</tr>
<tr>
<td>This work</td>
<td>Ultraviolet</td>
<td>Circular nano-disks</td>
<td>100</td>
<td>257.692 RIU$^{-1}$</td>
<td>1229.269</td>
</tr>
</tbody>
</table>

4. Conclusions

This paper introduces the use of the ultraviolet spectrum for refractometric biosensing applications. It draws inspiration from some of the latest research works on diamond metasurfaces for high-quality factor sensing in the ultraviolet spectrum. Therefore, a nano-sensor has been designed using diamond nanostructures and tested for the sensing of glucose through the ultraviolet spectrum. Comparison of this work with multiple other papers proves that the use of the ultraviolet spectrum remains a very less explored idea. The nano-sensor displays multiple resonance peaks that can be tracked for sensing analytes of different refractive indices. The lower wavelength peak corresponds to the formation of electric dipoles in the structure, while the higher wavelength shows the formation of magnetic dipoles. The lower and higher peak sensitivities are almost 67 nm/RIU and 100 nm/RIU, respectively. The Figure of Merit (FOM) for the 2 peaks is between 220 and 260 per RIU. The values of the Quality (Q) factors are as high as 1229. The novel achievement of this work is the spectral response containing dual sharp resonances, leading to high values of Figure of Merit (FOM) and Quality (Q) factor for sensing applications in the ultraviolet band. Future work can be carried out on optimizing all-dielectric diamond metasurfaces operating in the ultraviolet region to detect larger ranges of refractive indices, while having higher values of sensitivity for comparable quality factors.

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