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Role of presence of gold nanoparticles on enhancement of RF biosensing sensitivity

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Abstract

In this work, we explore the enhancement of sensitivity of sensing of biomolecules using Radio Frequency (RF) based sensing by utilizing gold particles. In particular, we investigate the impact of various parameters of gold particles such as shape, size and arrangements on enhancing sensitivity. The work uses a Microstrip Patch Antenna designed to resonate at 96.84 GHz. The sensor is facile, rapid and efficient and uses a sample volume of 17.5 nL. The results indicate that the presence of gold particles helps in improving the performance of RF sensors in detecting biomolecules. However, the extent of improvement depends on the properties of gold particles and is in the range of 30%–80% for the different cases studied in this work.

1. Introduction

In the last two decades, the scientific community has witnessed tremendous growth in the field of sensors. Sensors have been employed across diverse domains, including automobiles, hand-held electronic devices and numerous biochemical applications. They have revolutionized healthcare by facilitating the development of portable sensing technologies and Point of Care devices. There is high demand in the sensor market for devices that can monitor, sense and analyze analytes and parameters.

A major drawback associated with most conventional sensing technologies is that they require cumbersome and time-taking procedures [1–7]. Most require several cycles of labeling, incubation and washing and often, they cannot be conducted outside of sophisticated laboratories. In recent times, RF based sensing has featured as one of the suitable alternatives for developing such portable sensing technologies. They are characterized by several inherent advantages and have gained popularity by offering fast, facile and efficient solutions for biochemical applications.

RF based sensors have become popular due to the plethora of inherent advantages offered by them. They can highly simplify the sample preparation procedures and the sensing methodologies used by traditional techniques. Most of these processes are label-free and require minimum cycles of incubation and washing. A significant reduction in terms of reagents and the time required to perform the sensing are offered by these RF sensors, which can aid in developing rapid, simple and portable sensing techniques [8–10]. Another important advantage associated with them is that they require minute volumes of liquids for testing. For remote monitoring and testing, such a characteristic is highly favorable for developing Point of Care devices. RF-based sensors also have the advantage of being cheap and simple in terms of fabrication and integration procedures.

RF sensors essentially study the interaction of matter with EM waves. The response of these sensors changes in the presence of different samples due to the unique interactions taking place between the EM waves and the sample. They study the variation in different electrical and magnetic quantities such as permittivity, permeability and conductivity. Popular devices include Coplanar Waveguides, Interdigitated Capacitors, Cavity Resonators and Patch Antennas. These sensors are capable of performing both qualitative and quantitative analysis [8–11]. They have proven to be highly useful in performing comprehensive studies of a plethora of analytes. These

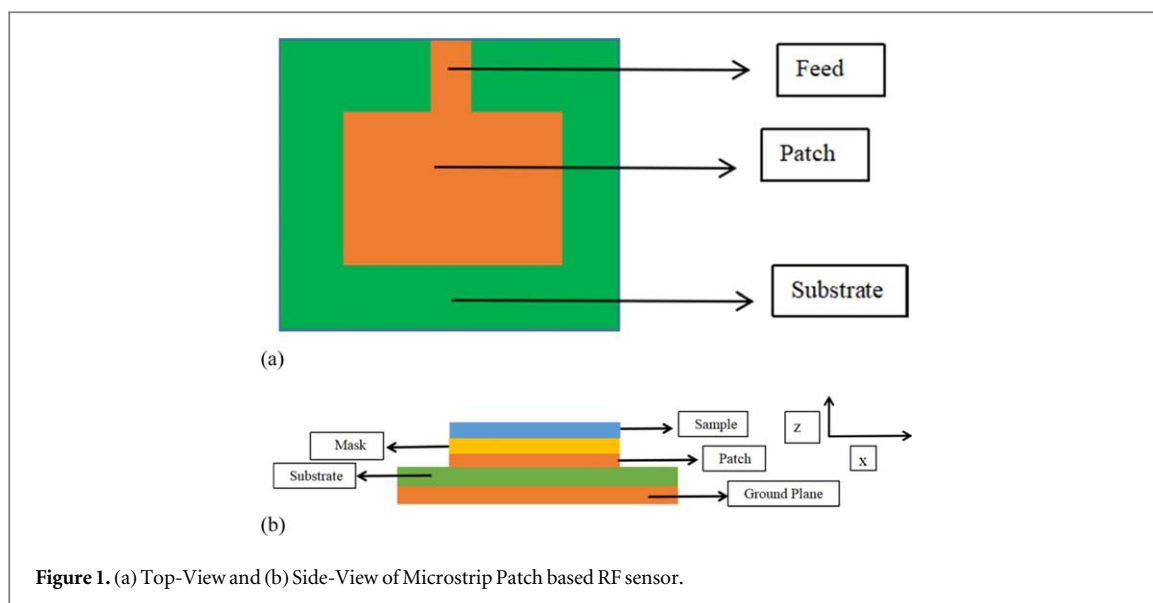


Figure 1. (a) Top-View and (b) Side-View of Microstrip Patch based RF sensor.

sensors have been exploited for diverse applications, especially in biochemistry and healthcare. The popular applications of RF based sensing include dielectric spectroscopy for the identification of bio-liquids [12–17], the study of the physiological and pathological state of cells, including tumor identification [18–24] as well as the detection of biomolecules and pathogens [25–29].

RF based sensing can also be explored for studying nanoparticles [30, 31]. The motivation behind performing such analyses is to see whether RF based sensing can detect variation occurring in the analytes in terms of concentration, arrangements, and morphology. Such analyses can help perform the primary characterization of nanoparticles remotely in the absence of sophisticated laboratory equipment.

However, there are certain challenges associated with the existing RF sensors. Most of these sensors face difficulties when trying to detect low concentrations of biochemical entities. There is a need to look for modalities that will enable easy detection of minute concentrations of samples. To this extent, this work explores the use of plasmonic particles [32–34], particularly gold nanoparticles, in enhancing the detection sensitivity.

In this work, RF based sensing is utilized to study aqueous solutions of nanoparticles and biomolecules. In particular, the detection of biomolecules in the presence of gold nanoparticles of different shapes, sizes and arrangements is studied. The motivation behind performing such an analysis is twofold. Firstly, such an analysis can help us understand how the different aspects of the analytes under test, such as shape, size, position and arrangements, get reflected in the characteristics studied under RF sensing. Secondly, it can help us assess whether any improvement in the efficiency of the detection of biomolecules takes place in the presence of gold nanoparticles. If such an improvement in efficiency is observed, it investigates how the efficiency varies with the variation of gold nanoparticles' different parameters.

The analysis is performed using the CAD tool, HFSS (High Frequency Structure Simulator). A microstrip-patch antenna based structure is designed in HFSS, and the different analytes are introduced in the sensing element to perform RF characterization. The S_{11} parameter is studied in terms of the resonance frequency, S_{11} and -3 dB bandwidth. A comprehensive understanding of the effect of the different characteristics of the nanoparticles and biomolecules is built using these parameters. In short, this work investigates the enhancement of efficiency of sensing of biomolecules using RF based sensors, in the presence of gold particles, by analyzing the variation in different aspects of the gold particles. The results can contribute towards the development of building efficient RF based sensing platforms.

2. Methodology

In this work, a microstrip patch antenna (figure 1) resonating at 96.84 GHz has been designed in HFSS. The antenna is designed on an FR4 substrate of thickness 160 μm . The copper patch on top is rectangular in nature with dimensions 0.5 mm and 0.7 mm and thickness of 0.0035 mm. The patch is fed using a microstrip feed of dimensions 0.05 mm width and 0.9 mm length. The ground plane is also built from copper of thickness 0.0035 mm. The entire patch structure is covered with an insulating cover of 0.001 mm thickness, to avoid degradation of the metal by the materials under test. The patch is covered with distilled water of volume 17.5 nL. The E-Field distribution of the patch-based structure in air is shown in figure 2.

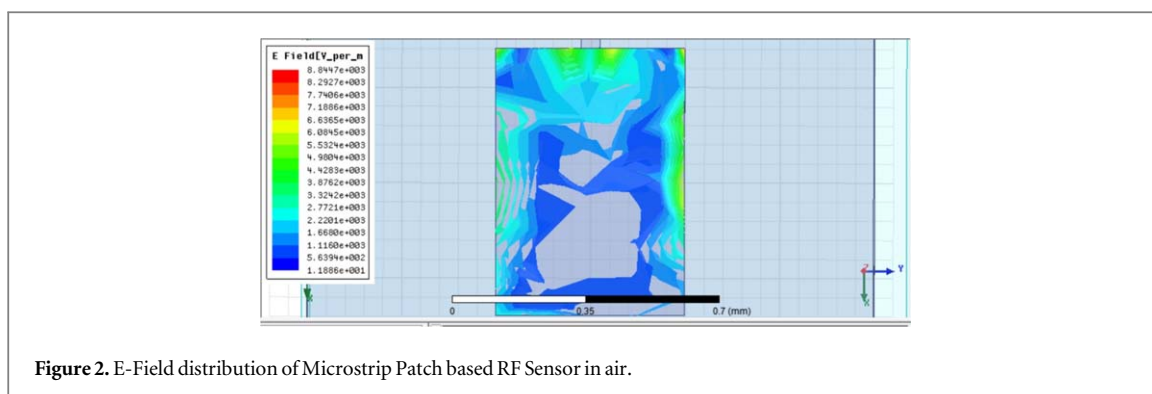


Figure 2. E-Field distribution of Microstrip Patch based RF Sensor in air.

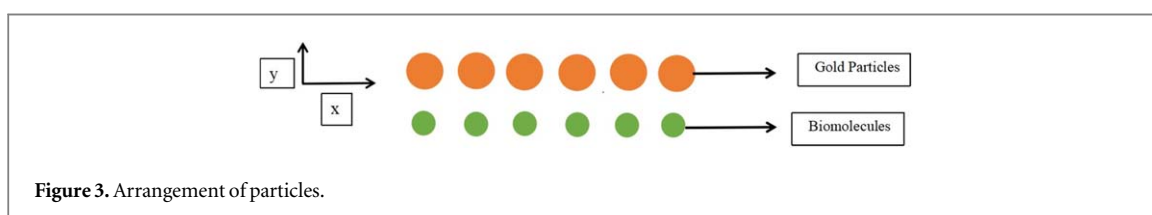


Figure 3. Arrangement of particles.

The S_{11} of the antenna structure is studied in the presence of different analytes and compared to perform analysis of the various MUTs (Material Under Test). In particular, the resonance frequency is measured. The resonance frequency of the different samples is compared to the resonance frequency obtained for the sensor loaded with distilled water ($\epsilon = 81$).

Large systems of biomolecules ($\epsilon = 4$) comprising 50–100 particles are studied in the presence of gold particles [39]. Various properties of the gold particles are varied, and their effect on the sensing performance is studied. The gold particles are kept close to the biomolecules and arranged as shown in figure 3.

The resonance frequency shift of the sensor in the presence of biomolecules is compared against the resonance frequency shift of gold-biomolecule systems. The percentage improvement in the same is used as a marker for sensitivity enhancement.

3. Results

Different parameters of the gold-biomolecule system have been studied. In particular, we have studied the variation of- (a) the size of the gold nanoparticles, (b) the distance between the gold and biomolecule, (c) the shape of the gold nanoparticle (d) the arrangement of the gold particles. The primary idea being explored is to understand (a) whether the variation in the various parameters of the gold nanoparticles help in improving the efficiency of sensing (b) if they do, how does it get reflected in the S_{11} response, and how can they be optimized to obtain maximal sensitivity.

3.1. Effect of distance between gold particle and biomolecule

The effect of the interparticle distance between the gold particle and the biomolecule is studied in this section using two distinct systems comprising particles of different sizes while the arrangement of gold particles and the biomolecules has been kept unaltered for the two cases.

The first system comprises spherical gold particles of radius 300 nm with biomolecules of radius 300 nm kept in their vicinity. The interparticle distance is varied from 1520 nm to 1660 nm in this case. The resonance frequency shift variation for this system is shown in figure 4. Similarly, we have taken disc-shaped gold particles of 100 nm radius and 10 nm thickness in the second system and kept in their vicinity biomolecules with an 80 nm radius. The particles are dispersed throughout the aqueous solution in a random fashion. The interparticle distance between the particles is varied from 10 nm to 100 nm. The resonance frequency variation for this system is given in figure 5.

Both cases (figures 4 and 5) indicate that as the distance between the gold particle and the biomolecule increases, the resonance frequency shift decreases. This could be attributed to the reduced interaction between the gold particles of different morphology and the biomolecules. For most of the cases studied in this section, the calculated resonance frequency shift is higher than that of the sensor with only biomolecules present. The enhancement in resonance frequency shift decays in an almost linear fashion as the distance increases, and for

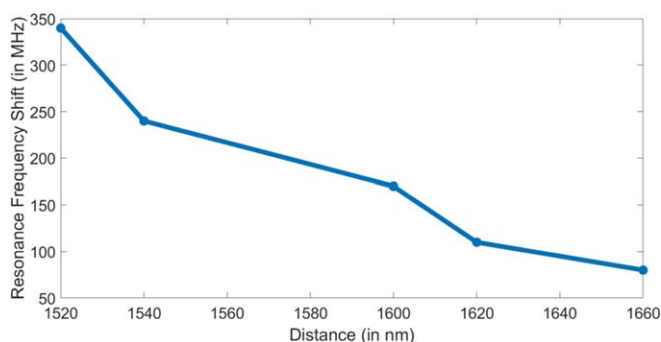


Figure 4. Variation of resonance frequency shift with distance between spherical gold particles and biomolecules.

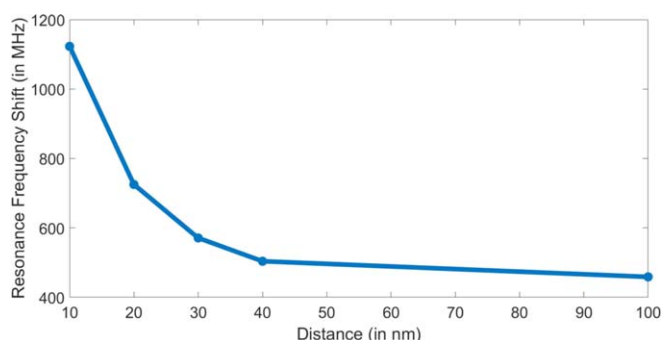


Figure 5. Variation of resonance frequency shift with distance between disc-shaped gold particles and biomolecules.

the larger distances studied for both instances, there is little to no enhancement observed. For instance, when the spherical gold particles are placed at a distance of 1520 nm, the resonance frequency shift is 340 MHz, which is 142% higher than in the case when only biomolecule is present. However, as the distance increases, this enhancement decays and at about a distance of 1700 nm, there is no enhancement present.

Similarly, in the case of the disc-shaped particles, we can see that as the distance between the gold and the biomolecules increases from 10 nm to 100 nm, the resonance frequency shift comes down from 1123 MHz to 459 MHz. The resonance frequency shift with only the biomolecules present is 724 MHz. Thus, in the presence of gold particles, as compared to the only biomolecules case, the resonance frequency gets enhanced by about 55% when the spacing is around 10 nm and results in no enhancement when the spacing reaches 100 nm.

The results suggest that while keeping gold particles in the vicinity of biomolecules can improve the efficiency of sensing by bringing forth a significant improvement in resonance frequency shift, the parameter is highly dependent on the actual distance between the two. For each system, this distance needs to be carefully evaluated to optimize the sensitivity enhancement.

3.2. Effect of shape of gold particle

As the previous section indicated, along with the distance between the particles, the size and shape of the gold particles play an essential role. In this section, we investigate the role of shape, and in the next, we study the role of the size of gold particles in enhancing sensitivity.

In order to study the effect of shape, we have two systems similar to the previous sub-section. In the first system, we have spherical biomolecules with 300 nm, kept in the vicinity of gold particles with a polygonal cross-section of radius 500 nm. In the second system, we have taken disc-shaped biomolecules of radius 80 nm and kept gold particles with a radius of 100 nm with polygonal cross-sections close to them. The number of sides of the polygonal cross-section is varied for both cases, and the effect of the same is studied on the enhancement of efficiency.

Both systems indicate that as the number of sides of the polygonal cross-section of the gold particles increases from 3 to 10, the resonance frequency shift obtained is higher. When the number of sides is varied for the gold particles with a 500 nm radius from five to ten, we see that the resonance frequency shift changes from 170 MHz to 300 MHz (figure 6), resulting in sensitivity enhancement in the range 21% to 114%.

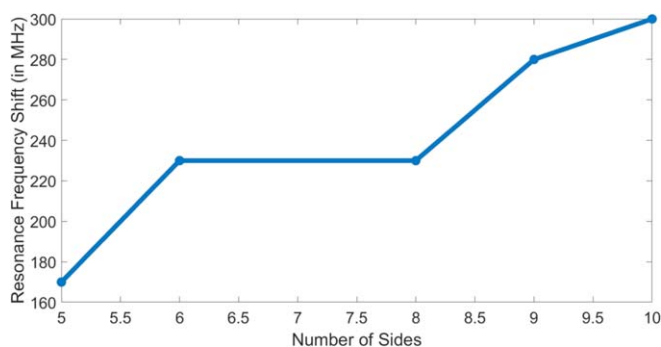


Figure 6. Variation of resonance frequency shift with number of sides of gold particles with polygonal cross-section, of thickness 150 nm and radius 500 nm.

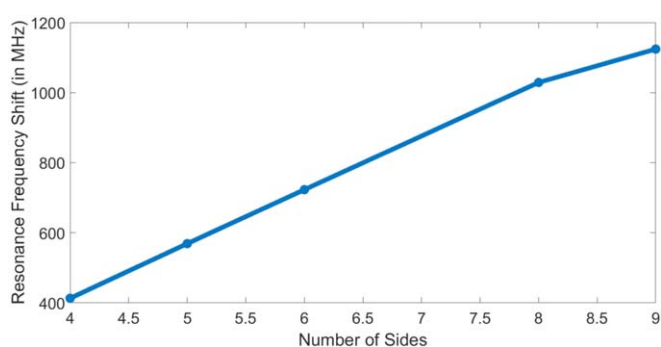


Figure 7. Variation of resonance frequency shift with number of sides of gold particles with polygonal cross-section, of thickness 10 nm and radius 100 nm.

When the number of the sides of the polygonal cross-section changes from four to seven for the gold particles with a radius of 100 nm (figure 7), we observe that the resonance frequency shift varies from 413 MHz to 1124 MHz. In the case of the square, pentagonal and hexagonal cross-sections, we see no enhancement of sensitivity. However, in the case of the other shapes, the sensitivity enhancement varies around 50%–60%.

These results help in further establishing that the increment in resonance frequency shift is a strong function of the shape of the gold particles. However, along with the interparticle distance and the shape, it is also required to place the biomolecules at favorable positions with respect to the gold particles. For example, positions around the sharp corners of the gold particles might help in delivering better efficiency.

3.3. Effect of size of gold particles

In this section, the effect of gold particles' size on the enhancement of resonance frequency shift is studied. The two systems used for performing the analysis in this section are the ones utilized in section-A.

When the spherical gold particles' radius is varied from 300 nm to 500 nm, the resonance frequency shift changes from 280 MHz to 580 MHz (figure 8). There is a dramatic improvement in the resonance frequency shift for each of these cases compared to the only biomolecule case. The sensitivity enhancement is in the range of 100%–300%.

Similarly, as the radius of the disc-shaped gold particles increases from 80 nm to 100 nm, the resonance frequency shift increases from 891 MHz to 1888 MHz (figure 9), resulting in significant improvement in the sensitivity.

The results indicate that, in general, irrespective of the shape of the gold particles, an increase in the gold particles' size is accompanied by an enhancement in the resonance frequency shift. Also, both sets of observations indicate that it is favorable to have gold particles bigger than the biomolecules to optimize efficiency. The results further demonstrate that the size of gold particles is a very important contender for deciding the sensitivity enhancement. However, the amount of shifts obtained varies by a large margin among the individual cases studied. Broadly, the improvement lies in the range of 28.57% to 74.24%.

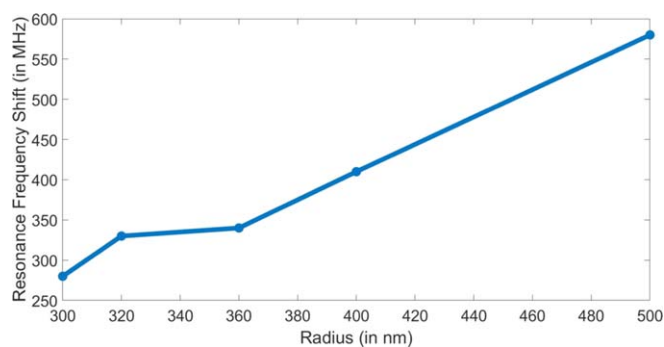


Figure 8. Variation of resonance frequency shift with radius from 300 nm to 500 nm of spherical gold particles.

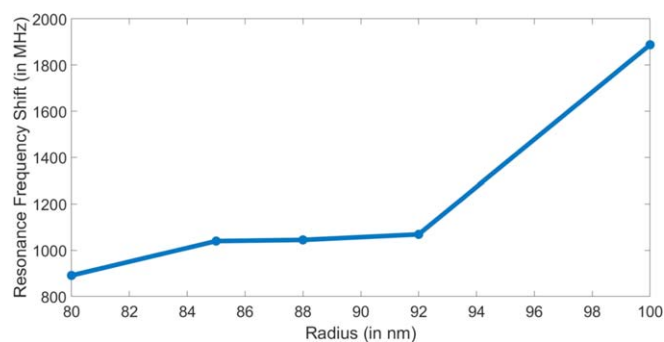


Figure 9. Variation of resonance frequency shift with radius from 80 nm to 100 nm of disc-shaped gold particles of thickness of 10 nm.

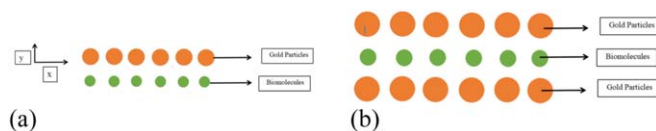


Figure 10. Arrangement of gold particles.

Table 1. Variation of S_{11} with arrangements of gold particles.

Distance (in mm)	No. of gold particles	Resonance frequency shift (in MHz)	S_{11} Am-plitude (in -dB)	-3 dB band- width (in MHz)
0.004	1	360	-30.163	430
	2	450	-38.047	170
0.0075	1	190	-31.165	480
	2	330	-34.187	270

3.4. Study of variation of arrangement of gold particles

This subsection explores the effect of variation of arrangements of gold particles on the improvement in the sensing performance. Spherical gold particles with a radius of 500 nm are taken and arranged in two different ways around the biomolecules, as shown in figure 10. For each of the two arrangements, two sets of interparticle distances are taken.

Table 1 indicates that when the interparticle distance is kept constant, an increase in the number of gold particles surrounding the biomolecules helps in increasing the efficiency of sensing. We can observe that for both the interparticle distance when the number of particles surrounding each biomolecule is two, higher resonance frequency shifts are obtained. In addition to that, we can notice a decrease in the S_{11} amplitude,

Table 2. Comparison of Design Complexity for Sensitivity Enhancement.

Reference no.	Sensitivity enhancement technique	Sensor design complexity
[35]	Use of Chaotic particle-ant colony algorithm	Very high
[36]	Use of interdigital capacitor and complementary split ring resonator	High
[37]	Use of integrated interdigital capacitor and symmetrical differential bridge-type inductor	Moderately high
[38]	Use of integrated interdigital capacitor and split ring resonator	High
This work	Use of gold nanoparticles loaded on a conventional microstrip patch	Low

accompanied by a decrease in the -3 dB bandwidth. The results indicate that, in general, it is favorable to have a higher concentration of gold particles compared to the biomolecules to obtain optimal sensing performance. The extent of the improvement will depend upon the other parameters discussed in the previous sections. Thus, it is imperative to ensure that sufficient interaction takes place between the biomolecules and the gold particles. In the absence of such interaction, no real improvement in efficiency might be observed.

3.5. Random arrangements of polydisperse gold nanoparticles

In order to simulate scenarios that resemble the complex experimental setups, simulations have been carried out to explore whether sensitivity enhancement is obtained when the various parameters of gold nanoparticles, such as their size, distance from biomolecules and their relative distribution in the solvent, are varied. The radius of disc-shaped gold nanoparticles varies from 80 nm to 100 nm, the interparticle distance varies from 230 nm to 330 nm, and the particles are randomly distributed in the aqueous solution. For the three cases studied, resonance frequency shifts of 893 MHz, 734 MHz and 947 MHz are obtained. In all the cases studied, the shifts are greater than the only biomolecule case, wherein a resonance frequency shift of 724 MHz is obtained. This further suggests that the presence of gold particles, even in a randomized fashion, can potentially contribute towards the enhancement of sensitivity. Also, it gives us a quantitative idea of the extent of variation that can be accommodated in gold nanoparticle-biomolecule systems to enable sensitivity enhancement. These results suggest that 25%–30% variability in the optimized parameter is tolerable and can still produce sensitivity enhancements.

4. Discussion

Different techniques have been adopted by RF sensing modalities to improve the sensitivity of detection. Primarily these techniques have opted for structural modifications in various ways to obtain optimal performance, as shown in Table 2. However, the requirement of structural modifications often imposes additional complexities on the design and fabrication process of the sensor. Such modifications are time-taking and require extensive computational resources. Often, these optimizations are performed by keeping a single analyte or several related analytes under consideration, which restricts the use of the sensor for diverse applications. In this work, an alternative methodology has been explored to utilize the use of gold nanoparticles to improve the sensitivity of detection. The nanoparticles can be tailored to fit the requirement of the sensing application, and no additional burden needs to be placed on the sensor design process. This would further make the sensor reusable for different analytes for multiple instances.

5. Conclusion

The results indicate that Radio Frequency based sensing can be effectively used to perform a comprehensive analysis of nanostructures. The sensing technique can differentiate between the variations in the various aspects of the gold nanoparticles. It further establishes that, in general, the presence of gold particles contributes to improving the sensitivity of detecting biomolecules. All gold particles' parameters, including shape, size, and position, play essential roles in ascertaining the extent of the enhancement obtained. The enhancement varies in the range of 30%–80%. In order to obtain maximal enhancement of sensitivity, there is a need to carefully monitor all of these parameters. A careful analysis of the morphology and the relative position of the gold particles is essential in building a comprehensive understanding of these systems. Thus, CAD-based exploration of such systems can help build efficient RF based sensing platforms that deliver optimal solutions.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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