

Improving Absorption Properties of Nanoparticles Using Self-Similar Array

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Abstract— In photo-thermal therapy, the localization of heat effects to cancer cells is an over growing approach in cancer treatment. This potentially reduces the side effects on tumor neighboring cells, improves the quality of life of the patient, and generally enhances the effectiveness of the hyperthermia. The optical properties of the nanoparticles depend mainly on their shape and size. The aim of this work is to enhance the photo thermal properties of noble metal nanostructures through improving their absorption characteristics. This is achieved using new shapes as well as new geometrical configurations of self-similar arrays of nanoparticles.

Keywords— *Quantum Dots (QDs); Nano-Particles; Localized Surface Plasmon Resonance (LSPR).*

I. INTRODUCTION

The incredible optical and electronic properties of nanocrystalline semiconductors were discovered in 1981 by A. Ekimov, who first synthesized nanocrystals embedded in a glass matrix [1]. Four years later, L. Brus, working at AT&T Bell Labs, performed the first colloidal semiconductor nano crystalline solutions [2, 3]. The optical properties of QDs occur in part due to a unique effect known as quantum confinement. Exciton is the phenomenon of generating electron hole pair in semiconductor materials which takes place when illuminating a semiconductor sample with a photon of efficient energy. Then, an electron of a QD can be excited from the valence band to the conduction band, leaving a positive hole in its place. Sometimes the QD radius is smaller than the exciton's Bohr radius. So, the confinement effects are strong, and the energy levels do not yet produce a continuous spectrum. As a result, the electronic and optical properties can be easily managed which are different from larger LED particles [4].

Nano-particles gained a wide reputation over the last years because of many reasons including the small size of nano-particles which helps in using them in different areas such as biomedical and pharmaceutical branches. Furthermore, they are cost effective and have high stability. On the other hand, gold nano-particles (AuNPs) were specifically used for drug delivery, antibacterial activities, cancer diagnosis, and imaging. Also, silver nano-particles (AgNPs) are used as agents in photothermal energy to remove cancer cells, bioimaging and catalytic agents in chemical reactions [5, 6]. Many shapes of

AuNPs have been developed to meet different therapeutic needs. In [7, 8], the absorption of light was determined by single elements of AuNPs. While in [9], different shapes of AuNPs were arranged in arrays which improved the absorption of light. The arrangement of AuNPs is the key part for the enhancement of light absorption. In this paper, we introduce a novel design which enhances the absorption of light by the AuNPs arranging them in a self-similar structure. Here, the nano-rod self-similar structure is better than the nano-sphere self-similar structure and the nano-sandwich self-similar structure because it offers the best absorption and therefore, it can be used for the therapy of tumors. The rest of the paper is organized as follows: Section II describes the process of exposing light to AuNPs. Section III explains the light scattering and absorption. The theoretical background of heating is explained in Section IV. Section V briefly illustrates the surface plasmon resonance (SPR). Section VI shows the description of the high frequency structure simulator (HFSS). The experimental procedure model is explained in Section VII. Simulation and results are presented and discussed in Section VIII. Section IX is devoted to the main conclusions of this study.

II. IMPROVED PROPERTIES OF GOLD NANO-PARTICLES

After exposing gold material to light, the following processes occur:

- *Absorption of light.*
- *Scattering of light at the same frequency as the incoming light (Mie or Rayleigh scattering).*
- *Absorbed light can be re-emitted (i.e., fluorescence)*
- *Improvement of the local electromagnetic field of the incoming light.*

In the case of AuNPs, all these processes are strongly improved. When light strikes a metal particle, a collective coherent oscillation of the free electrons of the metal particles of film appears due to the electromagnetic field of the photons with respect to the ions of the nano-particles along the direction of the electric field of the light. Depending on the characteristics of the metal film, the amplitude of oscillation reaches its highest value at a particular frequency, called surface plasmon resonance (SPR). The structure in case of nanometer range is called localized surface plasmon resonance (LSPR). The main difference between SPR and LSPR is that in case of LSPR the

oscillations are bounded in a small area. As a consequence, the electric field decays rapidly [10]. The LSPR spectrum form depends on the nanoparticle size, structure, distance between particles, properties of the nanoparticle material, and the properties of the surrounding medium [11].

III. SURFACE PLASMON ABSORPTION AND SCATTERING

The sum of absorption and scattering of electromagnetic waves after passing through a matter is known as the total light extinction. Using Mie theory, the surface plasmon absorption, scattering and total extinction efficiencies are studied [7], because for nanoparticles larger than 20 nm, the light absorption and scattering are described by taking into consideration all multiple oscillations [12]. Moreover, the optical absorption and scattering depend mainly on the size of the nanoparticles. Absorption is the dominant part of the total extinction in case of small size particles [13]. The calculations that are shown in Fig. 1 are performed using full Mie theory [14].

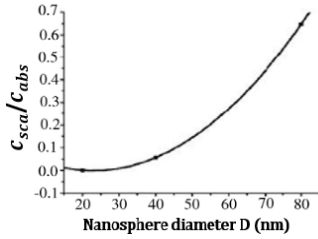


Fig. 1. Dependence of the ratio of the scattering to absorption cross-section on the diameter of gold nanoparticles [15]

As a result, the choice of AuNPs can be guided for biomedical applications. Larger nanoparticles are used for imaging due to the high scattering efficiency. Smaller nanoparticles are used for photo thermal therapy because light is mostly adsorbed by the particles and therefore is transformed to heat for cell and tissue destruction.

The optical tuning of AuNPs can be controlled by changing the particle shape, composition and structure. As predicted by Gan theory in 1915, transforming the AuNP from a sphere shape to a rod shape leads to the division of the SPR band into two bands: a strong band and a weak band. The strong band appears in the near infrared region. It refers to longitudinal band according to the electron oscillations along the long axis. On the other side, the weak band appears in the visible region at approximately 520 nm wavelength similar to that of gold nanospheres. It refers to a transversal band. By increasing the aspect ratio (length/width), the longitudinal band is red shifted largely from the visible to near-infrared region.

Furthermore, heat is produced efficiently from the absorbed light on a picoseconds time scale by quick electron-phonon and phonon-phonon interactions. The fast energy conversion and dissipation occurs due to SPR absorption. This results in heating the local environment by using light radiation with a frequency effectively overlapping with the nano-particles SPR absorption band. The strong efficient and localized light to heat conversion by gold nano-particles turns them beneficial for the photothermal therapy of diseases [16].

IV. THEORITICAL INVESTIGATION OF THE LIGHT-TO-HEAT CONVERSION

Gold nano-particles have superior light absorption efficiency. The electromagnetic radiation induces strong surface fields due to the coherent excitation of the electrons in the AuNPs. The rapid relaxation of these electrons generates strong localized heat capable of destroying surrounding targeted cancer cells via hyperthermia or other thermal-based effects.

For metal NPs, the amount of heat dissipated is higher than that dissipated in semiconductor NPs; due to an inter-band absorption process and the creation of a single mobile electron hole pair (exciton). In the non-existence of stage changing, temperature distribution around optically-stimulated NPs can be described by the equation of normal heat transfer [17]:

$$\rho(r)c(r)\frac{\partial T(r,t)}{\partial t} = \nabla K(r)\nabla T(r,t) + Q(r,t) \quad (1)$$

where r and t are the coordinate and time, $T(r, t)$ is the local temperature, and the material parameters $\rho(r)$, $c(r)$, and $K(r)$ are the mass density, specific heat, and thermal conductivity, respectively. The function $Q(r, t)$ depicts a source of energy coming from light dissipation in NPs:

$$Q(r, t) = (j(r, t) \cdot E(r, t))_t \quad (2)$$

where $j(r, t)$ is the current density and $E(r, t)$ is the stimulating electric field in the system. From Maxwell's equations, this field can be calculated.

For a group of metal and semiconductor NPs, Equation (1) should be solved numerically. But, for a single, spherical NP this equation can be easily solved analytically (using Fig. 2).

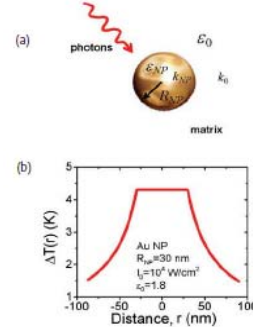


Fig. 2. (a) Schematic of an optically driven spherical nano-particle. (b) Calculated temperature increases for a single AuNP as a function of the distance from the NP center. The medium around the NP is water [17].

In the case of steady-state regime, the temperature around a single NP is described by a simple equation [17]:

$$\Delta T(r) = \frac{v_{NP}Q}{4\pi k_G r} \quad (3)$$

where r is the distance from the center of an NP, k is the thermal conductivity of the surrounding medium, and v_{NP} is the NP volume. Equation 3 is valid outside the NP, i.e., $r > R_{NP}$, where R_{NP} is the NP radius. The medium around NP can be water, a chemical solution, or a polymer.

Assuming that the wavelength of the incident light is much longer than the NP radius ($\lambda \gg R_{NP}$), one can also calculate analytically the heat generation Q .

$$Q = \frac{\omega}{8\pi} E_0^2 \left| \frac{3\epsilon_m}{2\epsilon_m + \epsilon_{NP}} \right|^2 IM(\epsilon_{NP}) \quad (4)$$

where E_0 is the amplitude of the incident radiation, ϵ_{NP} and ϵ_m are the dielectric constants of the NP and surrounding medium, respectively.

The maximum temperature increase occurs at $r = R_{NP}$ and is given by [18, 19]:

$$\Delta T_{max}(I_0) = \frac{R_{NP}^2 \omega}{3K_0 8\pi} \left| \frac{3\epsilon_0}{2\epsilon_0 + \epsilon_{NP}} \right|^2 IM \epsilon_{NP} \frac{8\pi I_0}{c\sqrt{\epsilon_0}} \quad (5)$$

where I_0 is light intensity inside the material. This equation shows the temperature increase with the NP size.

V. THEORIES CORRELATED WITH LSPR

In order to have an accurate representation of in-vivo blood dielectric properties, all factors are analyzed [20]. For a spherical metal nanoparticle with a size smaller than the wavelength of light, surface plasmon oscillation is dominated by the dipolar mode with a polarizability α given by [21]:

$$\alpha = 3\epsilon_0 V \left(\frac{\epsilon - \epsilon_m}{\epsilon + 2\epsilon_m} \right) \quad (6)$$

where V is the particle volume, ϵ_0 is the vacuum permittivity, $\epsilon(\omega) = \epsilon_r(\omega) + i\epsilon_i(\omega)$ is the complex frequency dependent dielectric function of the metal, ϵ_r and ϵ_i are the real and imaginary parts of the dielectric function of the metal nanoparticles, respectively, and ϵ_m is the dielectric constant of the surrounding medium.

The Mie solution to Maxwell's equations describes the scattering and absorption of incident light by spherical particles as mentioned in [22]. It represents a link for the extinction cross-section:

$$\sigma_{ext} = \sigma_{abs} + \sigma_{sca} \quad (7)$$

where σ_{ext} is the summation of the absorption cross-section and the scattering cross-section of the metal nanoparticles. For small particles ($d \ll \lambda$), the Mie solution is represented by:

$$\sigma_{ext} = 9 \frac{\omega}{c} \epsilon_m^3 / 2 v_0 \frac{\epsilon_i}{(\epsilon_r + 2\epsilon_m)^2 + \epsilon_i^2} \quad (8)$$

where $v_0 = (4\pi/3) R^3$ and ω is the angular frequency of the extinction radiation. The real and imaginary parts determine the wavelength position of the resonance and the bandwidth, respectively. As described in (8), the surface plasmon absorption band appears when $\epsilon_r = -2\epsilon_m$ if $\epsilon_i(\omega)$ is small or if it is only weakly dependent on ω . Consequently, the surface plasmon resonance (SPR) frequency is determined.

Changing shape of gold nanoparticles from spheres to rods results in the occurrence of a new SPR band at longer wavelengths, in addition to the plasmon band around 520 nm. In the case of particles of nonspherical shape, the resonance condition for surface plasmon oscillation is roughly achieved when:

$$\epsilon_r = -\left(\frac{1-L}{L}\right) \epsilon_m \quad (9)$$

where L is the depolarization factor. For the spherical shape, $L = 1/3$ but for anisotropic shapes, L has a unique value for each of the three axes X , Y , Z .

Depending on the characteristics of the metal film, the amplitude of oscillation reaches its highest value at a particular frequency, called surface plasmon resonance (SPR). The structure in case of nanometer range is called localized surface plasmon

resonance (LSPR). The main difference between SPR and LSPR is that in case of LSPR the oscillations are bounded in a small area. As a consequence, the electric field decays rapidly [10]. The LSPR spectrum form depends on the nanoparticle size, structure, distance between particles, properties of the nanoparticle material, and the surrounding medium [11].

VI. HIGH FREQUENCY STRUCTURE SIMULATOR (HFSS) DESCRIPTION

The HFSS originally stood for high frequency structure simulator. It is high performance 3D full wave electromagnetic field simulator. HFSS is based on frequency domain FEM (finite element method) simulation technology. It combines solid modeling, simulation and visualization in a common surrounding. Inside HFSS, Maxwell's equations are solved using finite element method.

The HFSS can be used to calculate parameters such as S parameters, resonance frequency, and fields. HFSS is commonly used in the design of antennas, filters, printed circuit board modeling and the design of complex radio frequency electronic circuit elements.

HFSS Analysis Design Flow [23]:

A. Pre-processing

It is necessary to define the problem space and its characteristics

- *Geometry construction*

The nanoparticles are arranged as self-similar structure pattern. The self-similar structure is exactly or approximately similar to a part of itself (i.e., the whole has the same shape as one or more of the parts).

- *Material assignment*

All the NPs in the simulation are made of gold. The AuNPs are immersed in blood.

- *Source/boundary assignment*

- a. Excitation is Wave Port (External)
- b. Surface Approximation is Radiation Surface
- c. Material Properties is Finite Conductivity of a conductor.

- *Solution 'setup'*

- a. Add Solution Setup
 1. Adapt Frequency (solution frequency: 600 THz)
 2. Adaptive solutions (maximum number of passes: 20 and maximum delta S: 0.02)
 3. Initial Mesh Options
 4. Adaptive Options
 5. Order Basis Functions (First order)
- b. Setup
 1. Sweep type (Fast)
 2. Frequency setup (Linear Count, Save Fields, Generate Fields (All Frequencies))

B. Solution

The actual solution of the problem defined in Pre-Processing above. Most of this step is 'automatic': Excitation Solution and *Meshing and Matrix Solution*

C. Post-processing

Evaluation of the results of the model (Plot reflection coefficient S_{11}).

VII. EXPERIMENTAL PROCEDURE MODEL

All the nano-particles used in the simulation are made of gold. To improve the absorption properties, the AuNPs are arranged in a self-similar structure pattern. The self-similar structure is exactly or approximately similar to a part of itself (i.e., the whole has the same shape as one or more of the parts).

The enhancement of absorption properties is important for cancer therapy. The AuNPs can be applied to patients by injection for medical treatments. The AuNPs can be prepared chemically [24]. The reflection coefficient S_{11} is measured by HFSS versus frequency. The minimum S_{11} occurs at the resonance frequency (plasmonic resonance) that represents the maximum energy transfer between the incident wave and the tissue (plasma surface).

VIII. SIMULATION RESULTS AND DISCUSSION

The high frequency structure simulator (HFSS) software is used to compare the performance of different shapes of nano-particles: nano-sphere, nano-rod, and nano-sandwich shape. Each of these shapes has been put in three different forms: single element, array elements and self-similar structure elements. The reflection coefficient, S_{11} is taken as a measure for the back scattering of the different structures.

Three tables (I, II and III) are shown in this section to compare between different shapes and different nano array structures and the corresponding reflection coefficients. This includes: the nano-rod, the nano-sphere and the nano-sandwich. Each element has been put in three forms: single element, array elements and self-similar structure elements.

Table I. Comparison between the optical properties of the single nano-rod element, nano-rod array elements and nano-rod self-similar structure elements

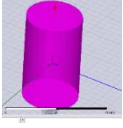
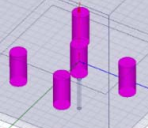
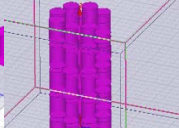
Shape	Nano-rod element [7, 9]	Nano-rod array elements [9]	Nano-rod self-similar structure elements (present work)
Design			
Dimension	Structure of the nano-rod element which consists of gold nano-rod (height=50 nm and radius=20 nm) inside dielectric substrate (has the dimensions 400×400×100 nm and $\epsilon_r = 58$).	Structure of the nano-rod element which consists of gold nano-rod (height=50 nm and radius=20 nm) inside dielectric substrate (has the dimensions 400×400×320 nm and $\epsilon_r = 58$).	Structure of the nano-rod element which consists of gold nano-rod (height=50 nm and radius=20 nm) inside dielectric substrate (has the dimensions 400×400×320 nm and $\epsilon_r = 58$).
S_{11} dB	-23.4 dB	-25.2 dB	-37.9 dB
Resonance frequency	367.4 THz	416.6 THz	572.3 THz
Shifted frequency		49.2 THz	204.9 THz

Table II. Comparison between the optical properties of the single nano-sphere element, nano-sphere array elements and nano-sphere self-similar structure elements

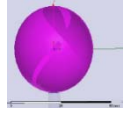
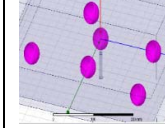
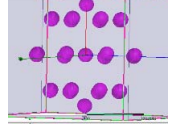
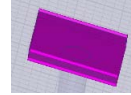


Shape	Nano-sphere element [7, 9]	Nano-sphere array elements [9]	Nano-sphere Self-similar structure elements (present work)
Design			
Dimension	Structure of the nano-sphere element which consists of gold nano-sphere (radius=20 nm) inside dielectric substrate (has the dimensions 400×400×60 nm and $\epsilon_r = 58$		Structure of the gold nano-sphere with 20 nm radius inside 400×400×340 nm and $\epsilon_r = 58$ dielectric substrate
S_{11} dB	-42.5 dB	-23.4 dB	-31.8 dB
Resonance frequency	1035.7 THz	768.4 THz	613.5 THz
Shifted frequency		267.3 THz	422.2 THz

Table III. Comparison between the optical properties of the single nano-sandwich element, nano-sandwich array elements and nano-sandwich self-similar structure elements

Shape	Nano-sandwich element [7, 9]	Nano-sandwich array elements [9]	Nano-sandwich Self-similar structure elements (present work)
Design			
Dimension	Nano-sandwich of two layers of gold sandwiched (each of 20×20×0.5 nm in-between by 20×20×1 nm glass as the dielectric material and $\epsilon_r = 5.5$ with 400×400×30 nm and $\epsilon_r = 58$ dielectric substrate		Nano-sandwich: two layers of gold sandwiched (each is 20×20×0.5 nm) in-between by 20×20×1 nm glass as the dielectric material and $\epsilon_r = 5.5$ and with 400×400×30 nm and $\epsilon_r = 58$ dielectric substrate
S_{11} dB	-12.3 dB	-15.1 dB	-23.3 dB
Resonance frequency	1293.5 THz	1389 THz	1211.5 THz
Shifted frequency		95.5 THz	82 THz

The effect of these designs results in:

- Shifting of the resonance frequency from that of the single element, the array elements and self-similar structure elements.
- Decreasing the reflection coefficient of some structures.

In [25], detailed theoretical as well as practical studies were conducted. They concluded that using short high power near infrared pulses for the irradiation of nanoparticles is better than long low power pulses in the process of localizing heat to the tumor location. They used both Arrhenius damage integral and the diffusive heat conduction equation to calculate the thermal

dose sufficient to kill the tumor keeping the normal tissues in healthy state. The minimum spread distance they achieved is 2.9 mm for short pulse high power treatment. In this study, we assumed that the arrayed particles are separated by a distance greater than the minimum spread distance of 2.9 mm. In the current study, we assumed that the separation distance between the array elements is greater than the minimum spread distance obtained in [25] to ensure that the accumulation of heat due to the array elements is linearly related to their number. In this case we can use low power elements to achieve the localization and the sufficient dose requirements keeping safe normal tissues. From Table II, it is noted that: in case of nano-sphere element, the highest absorption spectra occurs at $S_{11} = -42.5$ dB at a resonance frequency = 1035.7 THz, which is higher than the nano-sphere array and the nano-sphere self-similar structure elements. On the other hand, in Table I, the nano-rod self-similar structure elements give better results than the nano-rod and the nano-rod array elements. Also, in Table III, the nano-sandwich self-similar structure elements give better results than the nano-sandwich element and the nano-sandwich array elements. Figure 3 displays S_{11} of all single elements and shows that nano-sphere single element gives higher absorption spectra at $S_{11} = -42.5$ dB at a resonance frequency = 1035.7 THz.

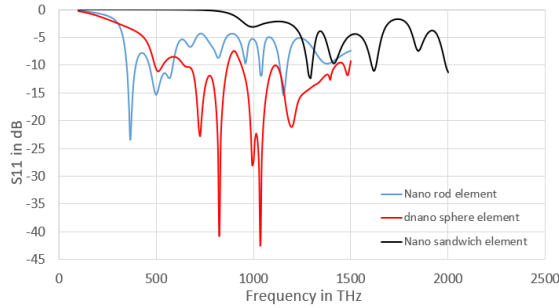


Fig. 3. The S_{11} reflection coefficient versus frequency for nano-sphere element, nano-rod element and nano-sandwich element.

Figure 4 shows S_{11} of all array elements, but in this case nano-sandwich array elements give higher absorption spectra at $S_{11} = -25.2$ dB at resonance frequency = 416.6 THz.

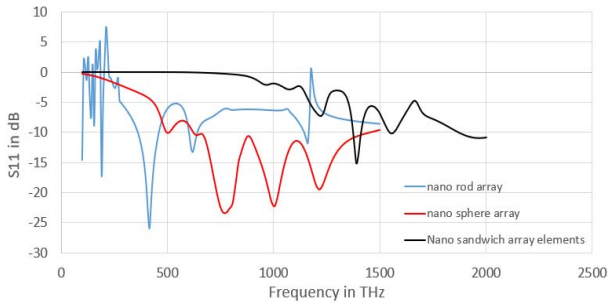


Fig. 4. The S_{11} reflection coefficient versus frequency for nano-sphere array, nano-rod array and nano-sandwich array.

Figure 5 displays S_{11} for all self-similar structure elements. From this figure, one can deduce that the self-similar structures of nano-sandwich is the best choice for diagnosis, while the nano-rod self-similar structure is the best choice for cancer tumors treatment which gives higher absorption spectra at $S_{11} = -37.9$ dB at a resonance frequency = 572.3 THz.

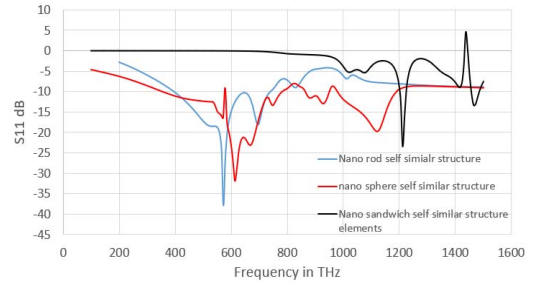


Fig. 5. The S_{11} reflection coefficient versus frequency for nano-sphere self-similar structure elements, nano-rod self-similar structure elements and nano-sandwich self-similar structure elements.

Comparing with previous work, better results are realized in this research. When using self-similar structure, better reflection coefficients are obtained. First, for nano-rod self-similar structure elements, nano-sandwich self-similar structure elements reflection coefficients are decreased to -37.9 dB and -23.3 dB, respectively. On the other hand, for nano-sphere self-similar structure elements reflection coefficient increased to -31.8 dB

Here, we focused on how AuNPs' size, shape, and surface modifications can influence the absorption properties. In addition, the absorption properties can be modified by changing the distance between the elements. For example, if we change the distance between the nano-spheres in the self-similar structure, the absorption coefficient can be increased if the interaction of waves is constructive, and decreases if the interaction is destructive. Also, in case of nano-rod self-similar and nano-sandwich self-similar structures, the absorption coefficient can be modified by changing the distance between the elements. Second, N. H. Ismail et al. [9] introduced a new technique using array elements, and obtained reflection coefficients of -25.2 dB, -23.4 dB and -15.1 dB for nano-rod array elements, nano-sphere array elements and nano-sandwich array elements, respectively. Our study using self-similar array leads to better results than the three different shapes introduced in [9]. From Fig. 3, it can be seen that the resonance frequency of the self-similar structure of nano spheres occurs at 613.5 THz which means that the resonance appears in the upper visible region. In Fig. 4, the resonance frequency of the self-similar structure of nano rods occurs at 572.3 THz. So, the structure of rod elements achieves $S_{11} = -37.9$ dB. On resonance appears also in the upper visible region. While in Fig. 5, the resonance frequency of the self-similar structure of nano sandwich occurs at 1211.5 THz, which means that the resonance appears in the ultra violet region. So, according to the application required, the shape, size and structure of AuNPs are chosen.

IX. CONCLUSION

Better understanding of the characteristics of NPs, LSPR and the setting around it means higher chances of success in the modern applications of nano-particles. Multiple shapes of nano-particles, nano-sphere, nano-rod and nano-sandwich are studied. In our work, NPs are sorted in an organized way, so, they would reach the optimum absorption spectrum. Further, the results of our work in case of nano-rod single element with absorption spectra at $S_{11} = -23.4$ dB at resonance frequency = 367.4 THz, are comparable with the results of Ref. [7] the designer of the nano-rod elements (in wavelength range 500-1100 nm). Also, it is found that self-similar the other hand, a novel shape nano-

sphere element is introduced. It has shown an optimum performance which overcomes the best results reported in the literature at $S_{11} = -42.5$ dB and frequency = 1035 THz. The obtained in a future work to determine temperature values in the medium surrounding the AuNPs [8]. This will be useful to determine if it can be used to destroy harmful cells without injuring the normal cells or not.

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