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Mixing rules and equation of effective permittivity of the medium having spherical inclusions in two different orientations of their basal planes for THz radiation emission

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Abstract
In this study, we determine the effective permittivity of a medium in which spherical inclusions are embedded in a host medium whose permittivity is given by ε_h. It is interesting to note that the inclusions are made up of graphite and their basal planes are oriented in two configurations, 1) parallel and, 2) perpendicular to the direction of the electric field of incident electromagnetic field. The quasi-static approximation is valid in the present study, i.e. the wavelength of the incident field in the medium is much higher than the scale of inhomogeneities and the medium can be treated as a homogeneous one. An easy to use and advanced equation has been derived with the said configuration following the Maxwell Garnett approach used to evaluate the resultant permittivity of the medium. The effective permittivity of the medium is then analyzed on the basis of fractional volume f_{t_i} of the inclusions in two different orientations along with two special cases when 1) $f_{t_i} \to 1$ and, 2) $f_{t_i} \to 0$. The role and nature of parallel and perpendicular orientations of basal planes on the effective permittivity is investigated. This method of calculating permittivity is useful for emission of THz radiation using laser-nanostructure interaction which is further helpful in the diagnosis and treatment of cancer diseases and in engineering fields related to material science.

Keywords
Permittivity, Inclusions, Fractional volume, Basal planes, Orientations .

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1. Introduction

The light-nanostructure interaction results in the interesting linear and nonlinear optical properties of the materials due to which it has gained importance in the past few years scientifically and technically [1]. While designing a material in composite material engineering problems, the required electrical and mechanical properties are kept in mind. The electrical behaviour of any material is described mainly by the conductivity and resistivity (reciprocal of conductivity) of the material and its dielectric strength. The dielectric behavior of any material is explained by the electrical permittivity. For material science and engineering applications, the value of the permittivity of the composite material should be high and to achieve required mechanical properties, a polymer matrix is required to form the composite [2]. The concept of homogenization has gained attention from past 25-30 years because of the increment in the practical needs for designing of novel composite materials and their applications to achieve the required frequency characteristics. Jylhä and Sihvola have obtained an equation for its applications in material designing in which overall permittivity of particle-filled composites is calculated [3]. The numerical validation using FDTD method for evaluating resultant permittivity of the mixtures has been performed by Kimmo Kalero Karkkainen et al. [4]. The same group has performed analysis using finite difference method on three-dimensional mixtures [5]. Rushman and Strivens have evaluated permittivity of two-phase systems [6] while for mixtures of anisotropic inclusions, the overall permittivity has been obtained by Sushko [7]. The effect of shape of the inclusion on the combined permittivity of isotropic or anisotropic medium has been investigated by Jones and Friedman [8]. A comparative study of the experimental results and results obtained by the mixing equation for evaluation of effective permittivity of composite media has been performed by Prasad and Prasad [9]. The effect of grain boundaries on graphite's dielectric behavior has also been studied by Xi and Chung [10].

The macroscopic properties of the medium are affected by the free parameters such as shape of the inclusions, individual phase and volume fraction of the inclusions. The fractional volume of the inclusions may vary from very dilute to high compositions [11]. The overall permittivity of the medium can be evaluated as a function of permittivities of the constituents and their fraction volumes. The inclusions, as well as host medium can be magnetic, dielectric, magneto-dielectric or conductive. Also, the inclusion material can be internally anisotropic or isotropic. To understand and predict the behavior of macroscopic permittivity of the medium, a theoretical background has been considered by Maxwell Garnett and Bruggeman in which they used a classical mixing approach to evaluate the effective permittivity of the heterogeneous mixtures [12]. 'The concept of homogenization' has been first introduced by Maxwell Garnett in 1904 and he developed a successful homogenization theory. It basically aims to simplify the complex electromagnetic medium with a homogeneous effective medium. For example, a colloidal solution having gold nanoparticles in water is approximated by homogenization theory and the permittivity of the medium is described in terms of the permittivity and fractional volume of the individual components of the complex medium [12]. The mixing formulae of both the Maxwell Garnett and Bruggeman work perfectly fine in the areas of remote sensing, microwave aquametry, material science and engineering etc. [3]. To check the validity of the approach, it is not an easy task to find an exact upper frequency limit since the exact response to the electromagnetic (EM) field of such a medium is beyond analysis. So, the concept of effective permittivity has its significance in the long wavelength limit only [13].

In the present study, we drive a simple and easy to use equation using Maxwell Garnett mixing approach for calculating the resultant permittivity of the medium having spherical inclusions in which basal planes are aligned in two distinct configurations to the direction of the electric field of incident electromagnetic radiation. The bound electrons of inclusions with permittivity ε_{t_i} are embedded in a background medium having permittivity ε_h. We consider the effects of fractional volume, radius of the inclusions and inter-particle distance between the inclusions, and distinct orientations of basal planes on the resultant permittivity of the medium.

2. Basic equations

We have taken spherical inclusions with two different orientations of their basal planes to the direction of incident electromagnetic field with fractional volume f and bound electrons permittivity ε_{t_i}. In hexagonal crystal structure, the basal planes are the planes parallel to the a_1,a_2 and a_3 axes but perpendicular to the symmetry axis. In our case, t_i is the symmetry axis perpendicular to the basal plane inside the spherical inclusion with distinct orientations. For better understanding of the picture let's consider that the incident electromagnetic field is propagating along z-axis and polarized along y-axis, i.e. $\mathbf{E} = E_0 \exp(-y/b)^2 \exp(i(kz-\omega t))\hat{y}$ then the electric field and basal planes are parallel when $(\mathbf{E} \perp t_{i=y})$ and they are perpendicular when $(\mathbf{E} \parallel t_{i=y})$. Using Maxwell Garnett approach, it is not required to specify about the propagation and polarization direction of the incident electromagnetic field as the resultant permittivity is not dependent upon the profile of the laser field. The propagation direction of Gaussian profile of

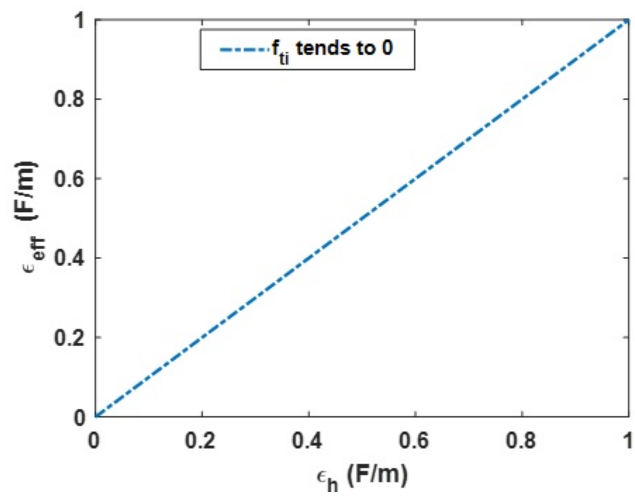

Figure 1. Variation of the effective permittivity with permittivity of host medium when number of inclusions vanishes.

the incident laser field along the z-axis and its polarization along the y-axis are considered as an example to visualize and comprehend easily. One can choose any kind of laser beam depending on the choice and requirement for the emission of THz radiation. The term effective permittivity comes in the expression of emitted THz field, calculated using wave equation when interaction between laser-nanostructure takes place via laser beating process. These spherical inclusions are mixed with medium with permittivity ε_h and fractional volume 1-f. It is important to mention that here inclusions have uniform permittivity throughout and we don't consider number of layers in the graphite inclusions.

With the help of classical mixing formulae, the effective permittivity of the said medium is estimated by assuming that the wavelength of the incident field in the medium is much higher than the scale of inhomogeneities. We first take one spherical inclusion with uniform permittivity ε_{t_i} either in parallel or perpendicular orientation of basal planes in a consistent background having permittivity ε_h. The term "consistent background" signifies that the medium in which the inclusions are embedded have constant permittivity throughout, which is given by ε_h.

The field $\mathbf{E}_e$ is the static external excited electric field which acts on the inclusion and $\mathbf{E}_i$ is the internal field induced inside the inclusion. It means the medium is excited by the field $\mathbf{E}_e$. The dipole moment $\mathbf{p}$ and polarizability α of a homogeneous spherically symmetric inclusion are related as $\mathbf{p} = \alpha \mathbf{E}_e$ where $\mathbf{E}_e$ is the field which acts on the inclusion [3]. The dipole moment is also directly proportional to the volume of the inclusion, the internal field inside the inclusion and contrast between the dielectrics of the environment and inclusion as [12], $\mathbf{p} = \int (\varepsilon_{t_i} - \varepsilon_h)\mathbf{E}_i dV$. The integration is limited only to the volume of the inclusion because $(\varepsilon_{t_i} - \varepsilon_h)$ is zero everywhere except inside the inclusion. The behavior of the inclusion with respect to the incoming electromagnetic field

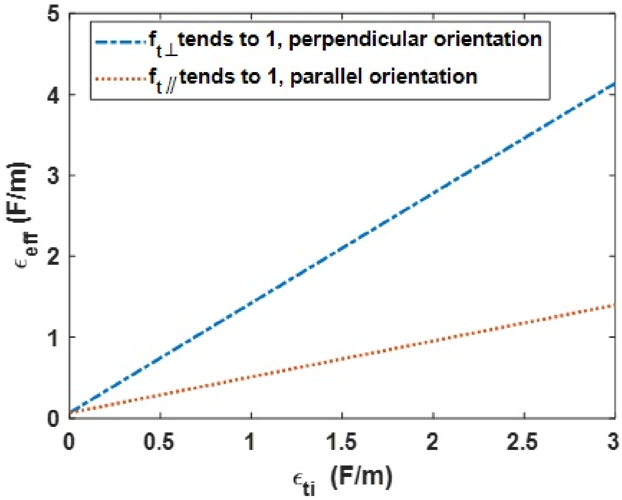

Figure 2. Variation of effective permittivity with permittivity of the inclusions in two different orientations of their basal planes in the absence of host medium.

can be measured by its polarizability. The relation between the static external field and internal field is written in the form [12], $\mathbf{E}_i = (3\varepsilon_h)/(\varepsilon_{t_i} + 2\varepsilon_h)\mathbf{E}_e$. Therefore, using this relation and expression of dipole moment, the polarizability can be written as, $\alpha = 3V\varepsilon_h(\varepsilon_{t_i} - \varepsilon_h)/(\varepsilon_{t_i} + 2\varepsilon_h)$, where $V = 4\pi r^3/3$ is the volume of spherical inclusion having radius r.
The polarizability we obtained is for one single spherical inclusion but for the case when more than one inclusions are present in the medium, then we need to consider the average electronic polarization density $\langle \mathbf{P} \rangle$ i.e. $\langle \mathbf{P} \rangle = n\mathbf{p}_{mix}$ where n is the number density of the inclusions and $\mathbf{p}_{mix}$ is the dipole moment of the inclusion in the mixture [13]. This $\mathbf{p}_{mix}$ is different from the $\mathbf{p}$ when the inclusion was in free environment i.e. when the inclusion was not affected by neighboring inclusions. In order to calculate the field that excites an inclusion, all other inclusions except the single one are replaced by average polarization which consistently surrounds the medium. So, $\mathbf{E}_L$ is now taken to be the exciting field whose existence is within the fictitious cavity of the shape of the inclusion. Therefore, the surrounding polarization adds to the exciting field of the inclusion which is given by, $\mathbf{E}_L = \langle \mathbf{E} \rangle + \langle \mathbf{P} \rangle / 3\varepsilon_h$ where $1/3$ represents the depolarizing factor in case of spherical inclusion [14] and the exciting field becomes higher than the average field. However, the formula , $\mathbf{p}_{mix} = \alpha \mathbf{E}_L$ is still valid because both $\mathbf{p}_{mix}$ and $\mathbf{E}_L$ are now higher in magnitude then obtained earlier. Hence, the average polarization can be written as $\langle \mathbf{P} \rangle = n\alpha \mathbf{E}_L$ and the classical equation for calculating the effective permittivity reads [12],

$$\frac{(\varepsilon_{eff} - \varepsilon_h)}{(\varepsilon_{eff} + 2\varepsilon_h)} = \frac{n\alpha}{3\varepsilon_h} \tag{1}$$

This equation contains the macroscopic quantities such as polarizability α and density of the inclusions n which are not convenient for macroscopic engineering. Therefore, it is advisable to obtain an equation in terms of components having defined permittivities in the medium. Such an equation is obtained using the expression of polarizability α and eqn. 1 simultaneously and can be written as [12],

$$\frac{(\varepsilon_{eff} - \varepsilon_h)}{(\varepsilon_{eff} + 2\varepsilon_h)} = f\frac{(\varepsilon_i - \varepsilon_h)}{(\varepsilon_i + 2\varepsilon_h)} \tag{2}$$

Here, $f = nV = 4\pi r^3/3d^3$ being the dimensionless quantity is the fractional volume of the inclusions in the medium, r and d being the radius of the inclusion and inter-particle distance between inclusions, respectively. On solving eqn. 2 algebraically, the effective permittivity of the medium reads [5],

$$\varepsilon_{eff} = \varepsilon_h + 3f\varepsilon_h \frac{\frac{\varepsilon_i - \varepsilon_h}{\varepsilon_i + 2\varepsilon_h}}{1 - f(\frac{\varepsilon_i - \varepsilon_h}{\varepsilon_i + 2\varepsilon_h})} \tag{3}$$

Here, when the number of inclusions vanishes i.e. $f \to 0$, effective permittivity reduces to permittivity of the host medium only. Also, when host medium vanishes i.e. $f \to 1$, overall permittivity is equal to permittivity of the inclusions.
The permittivity of the medium given by eqn. 3 is modified due to the presence of spherical inclusions with their basal planes oriented in two different configurations i.e. parallel and perpendicular with the direction to the incoming electromagnetic field [7] and is written in the form as,

$$\varepsilon_{eff} = \varepsilon_h + 3f_{t_\parallel}\varepsilon_h \frac{(\frac{\varepsilon_{t_\parallel} - \varepsilon_h}{\varepsilon_{t_\parallel} + 2\varepsilon_h})}{1 - f(\frac{\varepsilon_{t_\parallel} - \varepsilon_h}{\varepsilon_{t_\parallel} + 2\varepsilon_h})} + 3f_{t_\perp}\varepsilon_h \frac{(\frac{\varepsilon_{t_\perp} - \varepsilon_h}{\varepsilon_{t_\perp} + 2\varepsilon_h})}{1 - f(\frac{\varepsilon_{t_\perp} - \varepsilon_h}{\varepsilon_{t_\perp} + 2\varepsilon_h})} \tag{4}$$

Here, $\varepsilon_{t_\parallel}$ and ε_t are the permittivities of the inclusions when the basal planes are oriented parallel and perpendicular, respectively, to the direction of incident electromagnetic field. And, $f_{t_\parallel}$, $f_{t_\perp}$ are the volume fractions of the inclusions for the parallel and perpendicular orientations of the basal planes. The expression (4) is useful for applications in various fields, mainly for the emission of THz radiation using laser-nanostructure interaction [15–18], in engineering related to material science, when the number of inclusions are high [19, 20]. The method of calculating permittivity is useful for the emission of THz radiation using laser-nanostructure interaction. Metallic nanostructures have conduction electrons and bound electrons present in their outer and inner shells, respectively. The conduction electrons contribute to nonlinear current density when laser-nanostructure interaction takes place via laser beating process [15] while the contribution from bound electrons is calculated by evaluating the overall permittivity of the medium. Recent studies have been performed for the generation of THz radiation using a medium having graphite spherical and cylindrical NPs [16, 17]. Another study has been done on a medium containing conducting nano cylinders using hat-top laser beams for the emission of THz radiation [18]. By controlling the amount of shear flow on the nanoparticles, their orientation can be regulated over few

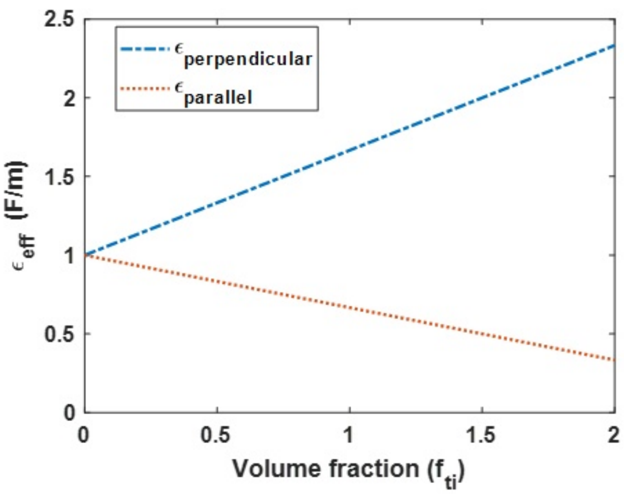

Figure 3. Variation of effective permittivity with fractional volume for permittivities of the inclusions in two different orientations of basal planes.

hundreds of micrometres. The rotation and translation of nanoparticles of arbitrary shape can be done by coupling the moulded elastomer with vision algorithm and feedback control system [21, 22]. Depending on the incident field profile, this may play role in other fields as well where the EM field interacts with matter [23–26] or charges are produced due to possible ionization of air [27–29].

3. Results and discussions

It is important to note that we consider our inclusions and background medium to be lossless, which implies that the value of the permittivity of the inclusions and host medium are taken be real only. These do not have any complex value. If losses are included in the inclusions, then the absorption of energy of EM wave takes place due to the imaginary part in the permittivity of the inclusion. We have taken argon (Ar) to be the host medium due to its nonpolar nature. The expression of resultant permittivity of the medium with two different orientations of the basal planes is given by eqn. (4). Now, we will see the changes in the effective permittivity of the medium with fractional volume of the inclusions in the parallel and perpendicular orientations f_{t_i}, permittivity of the host medium ε_h, permittivity of bound electrons in inclusions $\varepsilon_{t_\parallel}$ and $\varepsilon_{t_\perp}$. The variation of the effective permittivity given by eqn. 4 with permittivity of the host medium when the number of inclusions vanishes is shown in Fig.1. The graph shows linear dependence of ε_{eff} on ε_h i.e. as the host medium's permittivity increases, the effective permittivity of the medium also increases by the same amount. This is due to the fact that the effective permittivity of the medium is only dependent on the permittivity of the host medium when number of inclusions in both the orientations of the basal planes are absent in the medium. This can also be confirmed from eqn. 3 for the case when $f \to 0$ in which the orientations of the basal planes is not considered. The host medium can be a solid, liquid or a gas. It can be either conducting or dielectric. The host medium taken in this study is made up of argon (Ar) gas whose permittivity is 1.000574. So, the permittivity of the host medium is taken to be ~ 1. As a result, we obtain the linear behavior. So, it's the permittivity of the host medium which decides the overall permittivity of the taken medium, particularly for the case when inclusions are absent in the medium. Fikry et al. have investigated the effect of different host media like ethanol, PMMA in both liquid and solid phase on the intensity of fluorescence emission in both the liquid and solid phases of Rhodamine B [30].

Figure 2 shows the dependence of the overall permittivity with permittivity of the inclusions for the case when the medium is completely filled by the inclusions only in either orientation of their basal planes i.e. $f_{t_\parallel}$ and $f_{t_\perp}$. This is done for both the cases of perpendicular and parallel orientations. It is observed that the effective permittivity is higher for the perpendicular orientation of the basal planes than parallel orientation. This result obtained from the derived equation agrees with the experimental study performed by Taft and Philipp [31] in which they have obtained different values of permittivity, plasmon frequency and damping coefficient along the different orientations of basal planes due to the difference in the direction of the polarization of electron clouds along different directions upon irradiation by the laser field, which is due to the anisotropy in the NPs medium. Recent studies [15–18] have been performed on a medium having graphite spherical nanoparticles and nanocylinders for THz radiation emission. THz radiations of stronger intensity are found to be emitted when the basal planes are aligned parallel to the electric field of the incident field due to their lesser contribution towards effective permittivity. This result on lower effective permittivity is in agreement with the results obtained earlier [14, 17, 18].

Till now we have considered two cases of fractional volume i.e. 1) $f_{t_i} \to 0$ and 2) $f_{t_i} \to 1$. But, its value may vary from very dilute to high compositions [11]. So, the variation of the effective permittivity with fractional volume of the inclusions for different orientations of the basal planes is understood through Fig. 3. The trend is similar to the case $f_{t_i} \to 0$, as shown in Fig. 1, i.e. both the permittivities in parallel and perpendicular orientations start from $\varepsilon_{eff} = 1$ due to the presence of permittivity of the host medium at $f_{t_i} \to 0$. As the volume fraction approaches 1 in Fig. 3, the value of effective permittivity for the perpendicular orientation of basal planes is equal to 1.5 (F/m) and for parallel orientation of basal planes its value is ~ 0.6 (F/m); this also agrees with the results obtained in Fig. 2. The overall permittivity keeps on increasing linearly for the perpendicular orientation of the basal planes while nature is opposite for the parallel orientation i.e. it decreases for the higher values of the volume fraction. This decrease in the effective permittivity has been found to lead emission of THz radiations of much stronger intensity [17, 18]. The variation in the nature of the permittivity due to the parallel orientation of the basal planes has a little variation from linearity.

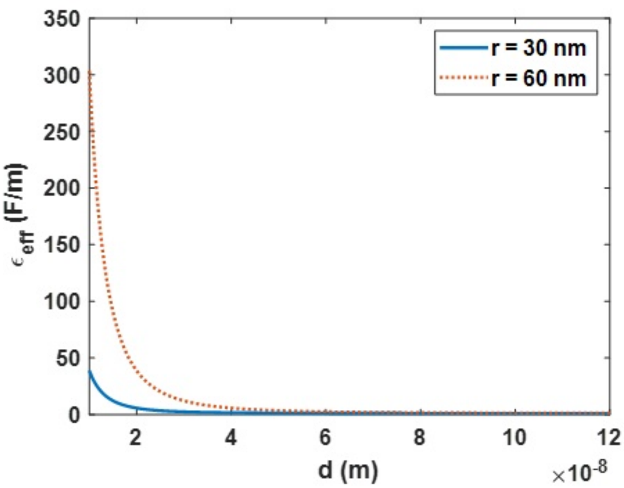

Figure 4. Variation of effective permittivity with inter-particle distance between inclusions for their different radii values.

In Fig. 4, we further explore the behavior of effective permittivity with the parameters of volume fraction i.e. radius of the inclusions and inter-particle distance between the inclusions. It can be observed that the value of effective permittivity is higher for the lower values of inter-particle distance between the inclusions and it approaches zero once the inter-particle distance is greater than 60nm. The value of effective permittivity is ~ 48 (F/m) when radius of the inclusion is 30 nm while its value increases to 6 times i.e. ~ 300 (F/m) when the radius of the inclusion is doubled i.e. 60 nm. This behavior is due to the linear dependence of effective permittivity with fractional volume of the inclusions. Therefore, the overall permittivity of the medium can be easily controlled by changing the values of the radius of the inclusions and inter-particle distance between them.
This combination of the graphite spherical inclusions and host medium that we have taken in our study is best suitable medium for the emission of efficient THz radiations from laser- nanostructure interaction with the help of laser beating process since the plasma frequency of graphite spherical inclusions lies in the THz range. The desired tuning of the frequency in THz range would be possible using the taken medium. The frequency tuning can be achieved in a way that longitudinal plasmon resonance peak and transverse resonance peak are obtained due to the resonant excitation of the medium when there is a matching of the beating frequency of the laser beams and plasmon frequency [17, 18]. The inclusions can be any shape like nanoparticles (NPs), nanocylinders (NCs), nanorods (NRs) etc. or a combination of two or three types of the nanostructures. This approach is novel in which two different orientations of the basal planes are included while calculating the permittivity of the medium which has not been considered in the past studies done till now. Using the calculations performed in this approach, the THz radiation generation using a medium having graphite nanoparticles in two different orientations of basal planes takes place via laser beating process. Also, it has been found that parallel oriented basal planes contribute much higher (10^4 times) to the THz radiation emission. Also, it has been found that the THz radiation of stronger amplitudes are emitted when greater radius of the NPs, lesser inter- particle distance between the NPs and lower magnitude of the effective permittivity of the medium are taken. In addition, the contribution to the emission of THz radiations has been found higher when only cylindrical NPs are present in the medium [16–18].

4. Conclusion

We have obtained a mixing equation using Maxwell Garnett approach that correctly predicts the effective permittivity for a medium that contains spherical inclusions with their basal planes aligned in two distinct orientations with the direction to the field of the incident electromagnetic field. We conclude that the effective permittivity is controlled by the radius of the NPs, inter-particle distance between the NPs, fractional volume and orientations of the basal planes. The value of the effective permittivity is higher for larger values of radius of the NPs, smaller inter-particle distance, higher fractional volume and for basal planes oriented in perpendicular direction. On one hand, the lower value of the effective permittivity for parallel oriented basal planes plays an important aspect for generating THz radiations of stronger intensity via laser-nanostructure interaction while the overall permittivity of the medium should be large in order to find applications in material science and engineering fields. The mixing equation for the effective permittivity for a medium like this is useful to modulate the frequency and amplitude of the emitted THz radiations which are further useful in the areas related to medical fields for diagnosis of cancer disease present in the human body.

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Conflict of interest statement:
The authors declare that they have no conflict of interest.

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