Course Name: Introduction to Microwave and Optical Metamaterials
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Week-4
Lecture-19

Lec 19: Mixing Rules and Effective Medium Theories

Hello, students. Welcome to lecture 19 of the online course on Introduction to Microwave and Optical Metamaterials. Today's lecture will be on mixing rules and effective medium theories. So, here is the lecture outline, we will continue where we left in the last lecture, we will metal dielectric composites and their mixing rules, classification of different engineered materials, mixing rules available there, introduction to effective medium theories and we will also look into the Maxwell-Garnett theory. So, in a composite where metal and dielectric components are intermingled with each other in a disordered manner, So, there applying the boundary conditions will become so complicated that determining the electromagnetic response of such a composite by solving the Maxwell's equation becomes practically impossible okay. Fortunately under certain ah conditions the solution can be signified significantly and that we will see today that for the study of optical properties of a composite system with inhomogeneity at a scale much smaller than the wavelength of interest okay.

Lecture Outline

- Metal-Dielectric Composites and Mixing Rules
- Classifications of Engineered Materials
- Mixing Rules for Engineered Materials
- Introduction to Effective Medium Theories
- Effective Medium Approach: Maxwell Garnett Theory

You can see that the electrodynamic scattering by individual metal or dielectric particles get overshadowed by the average response of the whole system. So, that allows you to do some kind of averaging, right? Therefore, we can investigate the optical properties of a microscopically heterogeneous composite by evaluating the effective dielectric function of the macroscopically uniform medium. So, that is very important. So, it has been underlined.

Metal-Dielectric Composites and Mixing Rules

- In a composite, where metal and dielectric components are intermingled with each other in a disordered manner:
  - The boundary conditions in the system are so complicated that the determination of its electromagnetic response by solving Maxwell's equations becomes practically impossible.
- Fortunately, under certain conditions the situation can be simplified significantly.
- For the study of the optical properties of a composite system with an inhomogeneity scale much smaller than the wavelength of interest:
  - Electrodynamic scattering by individual metal or dielectric particles is overshadowed by the average response of the whole system.

Source: W. Cai and V. Shalaev, Optical metamaterials, Springer US, 2011.

So we obtain this effective dielectric function in terms of the permittivity of the individual components as well as their respective volume fractions. So this method is known as the effective medium approach. Effective medium theories provide macroscopic models of inhomogeneous media based on analytical, numerical, and sometimes experimental techniques. So, let us go back and have a quick look at the classification of the engineered materials and that will allow us to understand where and in which situation this kind of averaging will be applicable. right so we have already seen this in our earlier lecture that engineered materials can be put into these four buckets right where you can purposely tailor their useful electromagnetic properties you have these ordinary materials you have these mixtures which are nothing but these ordinary materials getting combined to provide some Average properties.

Recap on Metal-Dielectric Composites and Mixing Rules

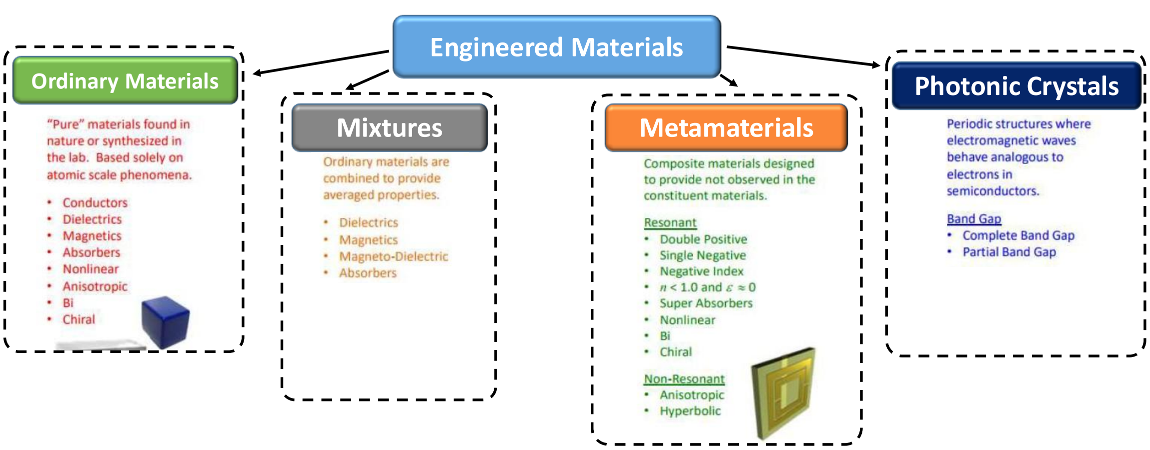
- Therefore, we can investigate the optical properties of a microscopically heterogeneous composite by evaluating the effective dielectric function of the macroscopically uniform medium.
- We obtain this effective dielectric function in terms of the permittivity of the individual components as well as their respective volume fractions.
- This method is known as the **Effective Medium Approach**.
- Effective medium theories provide **macroscopic models** of inhomogeneous media based on analytical, numerical, and sometimes experimental techniques.

Source: W. Cai and V. Shalaev, Optical metamaterials, Springer US, 2011.

So, you can have electrics, magnetics, absorbers, and magneto-dielectric in this category. We will also come to metamaterials where composite metamaterials, composite materials are designed to provide some properties which are never found in their constituent materials and we will also see this some examples here. Now, what are the mixing rules for engineered materials? So, let us say that we wish to mix multiple materials together to achieve some overall material property. So, it could be something like you have a material ε_1 and you have another material ε_2, you

Classifications of Engineered Materials

- As discussed in earlier lectures, Engineered materials are materials that are purposely tailored to exhibit useful and enabling electromagnetic properties.

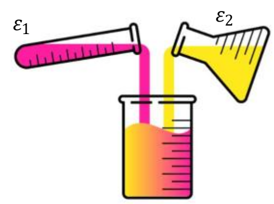


Source: W. Cai and V. Shalaev, Optical metamaterials, Springer US, 2011.

are mixing them together and you are expecting a new property which is ε_3. Now, the effective dielectric constant of this mixture can be written as ε effective.

Mixing Rules for Engineered Materials

- We may wish to mix multiple materials together to realize some overall material property.
- The effective dielectric constant of the mixture ε_{eff} depends on many things:
  - Shape of the particles
  - Size of the particles
  - Electromagnetic properties of the particles
  - Statistics on the particle distribution
  - Volume fill fraction of the constituent materials



ε_3=? (mixing rule)

Source: W. Cai and V. Shalaev, Optical metamaterials, Springer US, 2011.

That depends on several factors, first shape of the particles, size of the particles, electromagnetic properties of the particles and the statistics of the particle distribution and also the volume fill fraction of the constituent materials. Now that brings us to the effective medium theory approach. Now, a description of composite materials in terms of effective medium approximation becomes very valuable and it is a versatile tool to investigate, predict and design the electromagnetic response of natural and structured materials. So, effective medium models will equip the macroscopic Maxwell's equations with very simple constitutive relations. So, you can actually use those constitutive relations we have seen earlier, but here what is going to help you is that this effective medium approach will allow you to skip the complexity that comes from the light matter interaction happening at the constituent level.

Introduction of Effective Medium Theories

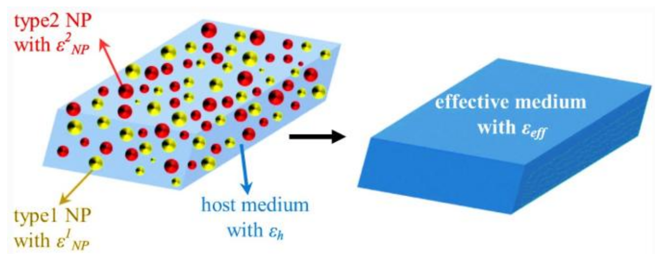
- A description of composite materials in terms of effective medium approximations is **a valuable and versatile** tool to investigate, predict, and design the electromagnetic response of natural and structured materials.
- Effective medium models **equip the macroscopic Maxwell's equations** with very simple constitutive relations, eliminating the complexity of simulating light-matter interactions at the constituents' level.
- When approaching an electromagnetic problem with an effective medium theory: **defining its limits of validity** is of extreme importance.
- Pushing any effective medium theory beyond these limits may lead to reasonable but only partially correct results or to wrong predictions.

Source: B. E. Saleh and M. C. Teich, Fundamentals of photonics, John Wiley & Sons, 2019.
Source: J. W. Haus, Fundamentals and applications of nanophotonics, Woodhead Publishing, 2016.

So, you do not need to go down to that microscopic level rather you can have a overall property of the material and then use the constitutive relation to describe the light matter interaction with that composite material. So, when approaching an electromagnetic problem with this effective medium theory it is very important to define the limit of the validity of this theory. So you have to understand that this theory cannot be applied blindly. So there is a range or there is a validity range / which this theory can give you a So, you can also find out what the maximum permittivity value is possible for the effective case. So, that is f ε r_1 + 1 - f ε r_2.

Introduction of Effective Medium Theories

- Effective medium models usually **depend on the electric and magnetic properties of the constituent materials**, the volume fraction of each constituent, and in some case the geometry of the structure at the constituent level.
- The fundamental limitation of these models:
  - Starting point of any approach for homogenizing structured materials is always the assumption that the wavelength of the field is much larger than the characteristic scale of the inhomogeneity.
- Depending on the size, permittivity, and permeability of the constituents, as well as the index of the hosting medium:
  - The limitations of the model may be more or less strict.



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Source: Z. Arefinia, Plasmonics, 15(6), pp.1661-1673, 2020.
Source: J. W. Haus, Fundamentals and applications of nanophotonics, Woodhead Publishing, 2016.

Now if it is a multi component system where there is a host and there are many many components. So, in that case, the minimum and the maximum can be written as this. So, 1 by ε min will be summation / m small m ranging from 1 to capital M. So, these are the number of components present in the host. So, you have to take the volume fraction of each component ok * the permittivity of that material ok and that is what we are doing here.

Okay, and you will see that all the volume fractions should add up to 1. Similarly, in the case of the maximum limit you will see you have σ small m ranging from 1 to capital M fm ε rm ok. So, that way, you can get the maximum refractive index, right? So, these equations are basically used as a first-order effective medium theory, okay. So, you can see that all these terms are the first-order terms. Now, if you take an example where your ε r_1 in a_2-component system is 200.

Effective Medium Theories: Weiner Bounds

- For mixtures, there exist **limits** on the range of possible effective permittivity values.
- **The Weiner bounds give the maximum and minimum values**.

Two Components Systems

$$\frac{1}{\varepsilon_{\min}} = f\frac{1}{\varepsilon_{\mathrm{r}1}} + (1-f)\frac{1}{\varepsilon_{\mathrm{r}2}}$$

$$\varepsilon_{\max} = f\varepsilon_{\mathrm{r}1} + (1-f)\varepsilon_{\mathrm{r}2}$$

Multiple Components Systems

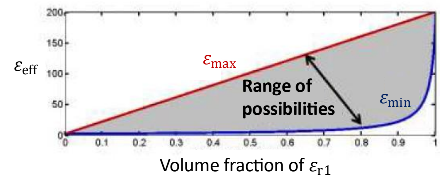
$$\frac{1}{\varepsilon_{\min}} = \sum_{m=1}^{M} \frac{f_{\mathrm{m}}}{\varepsilon_{\mathrm{r,m}}} \qquad 1 = \sum_{m=1}^{M} f_{\mathrm{m}}$$

$$\varepsilon_{\max} = \sum_{m=1}^{M} f_{\mathrm{m}}\varepsilon_{\mathrm{r,m}}$$

Note: These equations used for first-order effective medium theory.

f = volume filling fraction of the inclusion material ($\varepsilon_{\mathrm{r}1}$) in the medium ($\varepsilon_{\mathrm{r}2}$)

Example: For $\varepsilon_{\mathrm{r}1} = 200$, $\varepsilon_{\mathrm{r}2} = 2.5$



Source: Z. Arefinia, Plasmonics, 15(6), pp.1661-1673, 2020.
Source: J. W. Haus, Fundamentals and applications of nanophotonics, Woodhead Publishing, 2016.

So, you can say you have got a dielectric material of permittivity 200 embedded in a host medium of permittivity 2.5. So, in that case you can do this plot of volume fraction on the x axis and you have the effective permittivity on the y axis and you will see this is how the ε max will differ and this is how the ε mean will vary. So, you can actually have all these possibilities by mixing. Now, the two of the most commonly used effective medium approaches are this Maxwell Garnett theory and Bregman effective medium theory.

Effective Medium Theories

- Two of the most widely used effective medium approaches are the Maxwell–Garnett theory and the Bruggeman effective medium theory.
- Each of these two methods is based upon slightly different assumptions regarding the composite topology and the material properties of each constituent in the mixture.
- Depending on the relative concentration of the inclusions and the process of fabrication, metal-dielectric composites may have different types of microscopic structures.
- To present this point more clearly, TEM images of two samples with typical topologies are shown in Fig:

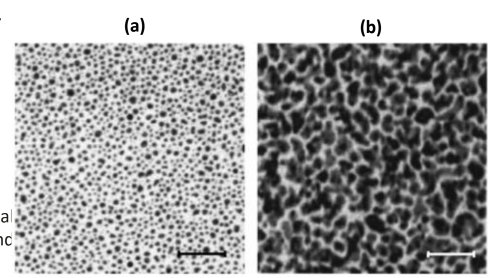


Figure. Transmission Electron Microscopy (TEM) images of typical metal-dielectric composites in (**a**) the Maxwell–Garnett geometry and (**b**) the Bruggeman geometry.

Scale bar is 200 nm for both images.

Source: W. Cai and V. Shalaev, Optical metamaterials, Springer US, 2011.

So, these are the two most important ones. So, each of these two methods basically depend on slightly different assumptions regarding the composite topology and the material properties of each component in the mixture. So we'll take them up one by one. Now, depending on the relative concentration of the inclusions and the process of fabrication, metal dielectric composites may have different types of microscopic structures. So, here you can see the two different cases, okay? So, we are presenting here TEM transmission electron microscopy images of two samples which have two different topologies.

Effective Medium Theories

- In the first composite sample, the inclusions particles (the black areas of the image) embedded in the host material (the white areas of the image) are quite dilute and have well-defined, spherical shapes.
- This is usually called the **Maxwell–Garnett geometry**.
- When the two constituent materials intermingle with each other and the two materials play symmetric roles, as shown in Fig. b, it is difficult to say which is the host and which is the inclusion.
- This type of topology is commonly referred to as the **Bruggeman geometry**.

(a) **(b)**

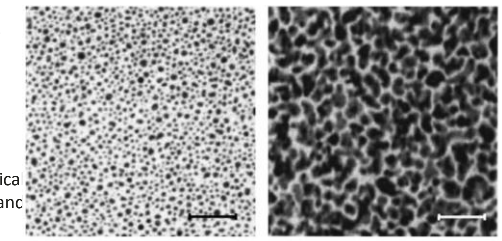


Figure. Transmission Electron Microscopy (TEM) images of typical metal-dielectric composites in (**a**) the Maxwell–Garnett geometry and (**b**) the Bruggeman geometry.

Scale bar is 200 nm for both images.

IIT Guwahati | NPTEL swayam Source: W. Cai and V. Shalaev, Optical metamaterials, Springer US, 2011.

So, you can see here that these are both metallic dielectric composites. The first one is basically showing Maxwell Garnett geometry, and the other one is showing, you know, the Brugman geometry. So, just to tell you the dark and the bright areas represent the dark one represents metal and the bright areas represent dielectric components. Now, if you carefully look into the first sample / here, it looks like the inclusion particles which are basically the black areas in the image, they are embedded in the host material which is the white area right. So, you can see that these things are quite dilute and all the particles have well defined spherical shape few large few small, but does not matter the shape is very well defined.

Effective Medium Theories: Maxwell Garnett Theory

- This is the ***classical approach for homogenizing media*** with small inclusions dispersed in a continuous host medium or matrix.
- The basic structure is a two-phase medium with separated grains of a guest material, the inclusions with relative permittivity ε_i, hosted by a background medium (host) with relative permittivity ε_h.
- *We restrict the analysis to the case of nonmagnetic and isotropic materials.*

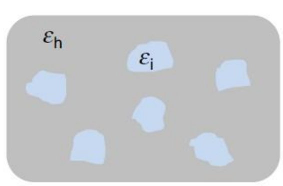


Figure: Illustration of Maxwell Garnett homogenization.

IIT Guwahati | NPTEL swayam Source: W. Cai and V. Shalaev, Optical metamaterials, Springer US, 2011.

So, you can typically call this as a Maxwell Garnett geometry. On the other hand if you look into the other image here you see that the two constituent materials are basically intermingled with each other. And the two materials, the dark and the light, or the bright areas, these two materials basically play symmetric roles. So, it is difficult to say which is the host and which is the inclusion. And this kind of geometry is typically referred to as Bragman geometry.

Effective Medium Theories: Maxwell Garnett Theory

- If the inclusions are small enough, then a quasi-static approximation can be adopted.
- For positive permittivity inclusions the following rule of thumb is considered to be conservative: **the particle size should not exceed one-tenth of the effective wavelength**, which is the wavelength measured in the effective medium.
- However, in case of metallic or negative-permittivity inclusions, the limits of validity may be stricter, especially near the localized surface plasmon resonances.

Figure: Illustration of Maxwell Garnett homogenization.

IIT Guwahati | NPTEL swayam Source: W. Cai and V. Shalaev, Optical metamaterials, Springer US, 2011.

Now, this is the classical approach for homogenizing media which has got small inclusion dispersed in a continuous host medium or matrix and this is typically the case with Maxwell-Garnett theory. So, you have got a host which has got a refractive index of ε sorry relative permittivity of ε h and it has got a inclusion which has got a relative permittivity of ε i and these are all separate grains ok distinct separate grains. So, in that case ok, if we restrict our analysis to the case of non magnetic and isotropic material and if we assume that these inclusions are much smaller in size as compared to the wavelength, you can take up a quasi static approximation for finding out the overall permittivity of this medium. So, for positive permittivity inclusions that means, if these inclusions are made of dielectric material ok, you can use a rule of thumb ok that the particle size should not exceed λ by 10.

Okay. That means and that λ by 10 the λ is basically the wavelength measured in the effective medium ok. So, that is the kind of rule of thumb. However, if you have metallic or negative permittivity inclusions the limit of this validity becomes much stricter because those metallic nanoparticles may have resonance at your particular wavelength of interest. And when the incident light is close to the resonance wavelength, the validity limit will become much stricter. So, in the absence of any information about the shape of the inclusions ok, it is very common and it is a natural approach to assume that these are all tiny spheres ok.

Effective Medium Theories: Maxwell Garnett Theory

- In the absence of any information about the shape of the inclusions, the most natural approach is to assume that they are small spheres.
- The idea behind the Maxwell Garnett homogenization approach is exemplified in the **Figure**.
- If the material is excited by an external electric field $(\mathbf{E}_e)$ in the quasi-static approximation such a field can be considered to be constant on the length scale of each sphere.

Figure: Illustration of Maxwell Garnett homogenization.

IIT Guwahati | NPTEL swayam Source: W. Cai and V. Shalaev, Optical metamaterials, Springer US, 2011.

And will show you the idea behind this Maxwell Garnett homogenization approach with a figure that if you again got a material host material with permittivity ε h which is got tiny in inclusions of relative permittivity ε i. And if this material is excited by an electric field ok the first thing would be to consider that in quasi static approximation the field is assumed to be constant / the length scale of each sphere ok. That means the spheres need to be sub wavelength much much smaller than the wavelength that means / this volume of the sphere there will be no significant special variation of the electric field. So, in such situation you can always represent this entire system with a effective permittivity and that can be written as ε MG Maxwell Garnett. So, if you go in details you will see that on the left the microstructure that you are discussing right now is typically a two phase medium with very small guest inclusions of unknown shapes and dielectric constant which is dispersed.

Effective Medium Theories: Maxwell Garnett Theory

- On the left is the microstructure under investigation, which is a two-phase medium with very small guest inclusions of unknown shapes and dielectric constant ε_i dispersed in a continuous host medium with permittivity ε_h.
- On the central panel is the Maxwell Garnett "view" of the material on the left, in which the inclusions are described as small spheres of dielectric constant ε_i.
- On the right is Maxwell Garnett homogenization, with effective permittivity ε_{MG}.

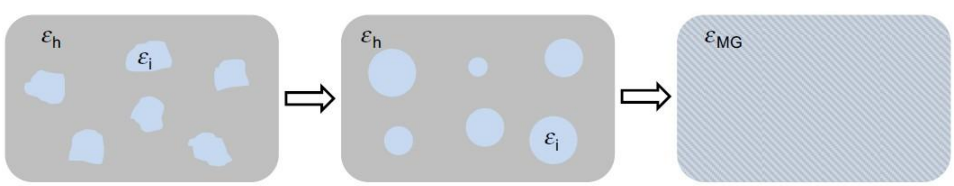


Figure: Illustration of Maxwell Garnett homogenization.

IIT Guwahati | NPTEL swayam Source: W. Cai and V. Shalaev, Optical metamaterials, Springer US, 2011.

okay and this has got a dielectric constant ε i dispersed in a continuous host medium which has got a permittivity of ε h. This tells you the Maxwell Garnett view of this particular material where each inclusion can be considered as a small sphere again with the permittivity of ε i and here you will see how the Maxwell Garnett homogenization looks like with permittivity effective permittivity of ε MG. So, this overall system can be now represented as a homogeneous material with refractive index of ε mg. So, let us first focus on the response of each isolated sphere to the excitation. Now as I mentioned that if you are considering the spheres to be very small it will act as a point source with an electric dipole moment which is proportional to the electric field or in other words you can write the response of an isolated sphere in a Host medium in terms of its polarization, which is coming from OK.

Effective Medium Theories: Maxwell Garnett Theory

- First we focus on the response of each isolated sphere to this excitation.
- Since the sphere is very small, it acts as a point source with an electric dipole moment proportional to the applied field.
- In other words, the response of an isolated sphere in the host medium is: $\mathbf{p}_h = \varepsilon_0 \varepsilon_h \alpha \mathbf{E}_e$

  where ε_0 is the vacuum permittivity, $\alpha = 3V \frac{\varepsilon_i - \varepsilon_h}{\varepsilon_i + 2\varepsilon_h}$ is the static electric polarizability of the sphere, and V is the sphere's volume.

Figure: Illustration of Maxwell Garnett homogenization.

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Source: W. Cai and V.Shalaev, Optical metamaterials, Springer US, 2011.

So, in terms of its so, in other words you can the response of an isolated sphere in the host medium can be written as $\mathbf{p}_h = \varepsilon_0 \varepsilon_h \alpha \mathbf{E}_e$. So, here you will see that ε_0 is basically the vacuum permittivity α is basically giving you the static electric polarizability of the sphere that depends on the volume of the sphere also on the relative permittivity of the material and the host medium ok. So, this is basically giving you the electric dipole moment, which is proportional to the incident electric field. So, this proportionality constant is known as electric polarizability. Now the field inside the sphere can be written as $\mathbf{E}_i = \frac{3\varepsilon_h}{\varepsilon_i + 2\varepsilon_h} \mathbf{E}_e$ that is basically the external electric field.

So, the field inside is uniform, and it is parallel to the external electric field. So, what you can see is that the polarizability of the sphere is isotropic. Because you have considered the permittivity and the shape of the inclusion to be isotropic as well. So, the next step is to create an effective model of this distribution of small spheres. So, you are now trying to move from here to there.

Effective Medium Theories: Maxwell Garnett Theory

- The field inside the sphere, $\mathbf{E}_i = \dfrac{3\varepsilon_h}{\varepsilon_i + 2\varepsilon_h}\mathbf{E}_e$,is uniform and parallel to the external field $\mathbf{E}_e$.
- The polarizability of the sphere is isotropic since both the permittivity and shape of the inclusions are assumed to be isotropic.
- The next step is to create an effective model of the distribution of small spheres (**transition from the center to the right panel in Figure**).

Figure: Illustration of Maxwell Garnett homogenization.

IIT Guwahati | NPTEL swayam   Source: W. Cai and V. Shalaev, Optical metamaterials, Springer US, 2011.

So, we understood that how we move from this to that we have to presented contribution of each point dipoles in terms of the dipole moments and polarizability of the spheres ok. And now, once the spheres are basically reduced to all-electric point dipoles, okay. Now you have to understand that how the field radiated by each point dipole is getting influenced in the presence of all other dipoles ok. That means you have to take into consideration the number of dipoles that is present in a unit volume and say you have n number of point dipoles like that. So, in that case you have to think of that the effective permittivity that you are getting is nothing but an average or microscopic constitutive relation that is coming to link the average electric field to the average displacement field D.

Effective Medium Theories: Maxwell Garnett Theory

- The spheres are reduced to electric point dipoles, and the field radiated by each dipole is now influenced by the presence of all the other dipoles.
- At this point the information required is the number of dipoles per unit volume, N.
- The definition of **the effective permittivity** is based on the average, or macroscopic, constitutive relation that links the average electric field $\langle \mathbf{E} \rangle$ to the average displacement field $\langle \mathbf{D} \rangle$.

Figure: Illustration of Maxwell Garnett homogenization.

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So, the average electric field can be represented like this okay and the average displacement field can be represented like this. So, you are basically finding out this effective permittivity that will be correlating this effective or average displacement field with average electric field. Now, the average parameter integrates / sufficiently large volume in order to provide an accurate description of the field or the average field in the original medium. So, one can write this expression as: The average of the displacement field will be equal to ε_0 ε MG * the average electric field. So, this is the effective relative Maxwell Garnett permittivity that basically models the original mixture.

Effective Medium Theories: Maxwell Garnett Theory

- The average operator integrates over sufficiently large volumes in order to provide an accurate description of average fields in the original medium. Hence, one can write:

$$\langle \mathbf{D} \rangle = \varepsilon_0 \varepsilon_{MG} \langle \mathbf{E} \rangle$$

where ε_{MG} is the effective (relative) Maxwell Garnett permittivity that models the original mixture, as shown in Figure.

- One can also see the average medium response as the average response of the host medium plus the average response of the dipoles, so that

$$\langle \mathbf{D} \rangle = \varepsilon_0 \varepsilon_h \langle \mathbf{E} \rangle + \langle \mathbf{P} \rangle$$

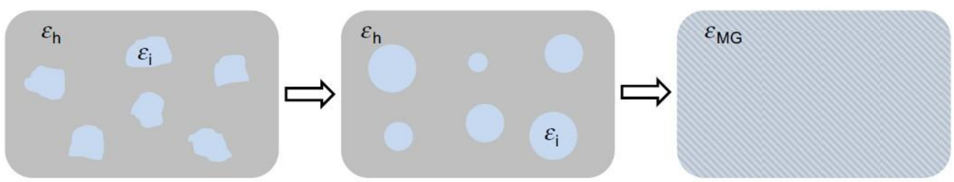

Figure: Illustration of Maxwell Garnett homogenization.

So, one can also see that the average medium response ok that you are getting is basically nothing, but the average response from the host medium ok. + you are basically getting the response from the dipoles coming from the small spheres which are nothing but the small inclusions that is there in the host medium and that is finally giving you the effective permittivity. So, we look into the equations and how we finally calculate. So, we have already got the idea that this value, the effective permittivity, has to be within the Wiener bounds. So, there is a limit of the maximum and the minimum value of the permittivity possible based on the individual values of the host medium and the inclusions and also their volume fraction.

So, the mixture will have something in between. So, here what one thing is important that the inclusions need to be of sub wavelength size and then what you are finding out is that first you consider each of these inclusions ok and then you assume them as point dipoles and then because this point dipole in the in an oscillating electric field will also be oscillating and they will radiate and the the scattered field from this dipoles will be kind of interacting with the other dipoles in the vicinity and then you will get a overall response from the material and that is what this particular theory is helping you to approximate, right. with that we will stop here and we will look into the equations and how we can derive this Maxwell Garnett theory and also we will also move and discuss the Bragmann theory in the next lecture. So, if you have got any question regarding this particular topic you can drop an email to this email address mentioning the course name and the lecture number on the subject line.

Thank You