

Left Handed Maxwell Systems In Optical Regime

PART-3

Effective Medium Approximations for Optical Meta-Material

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Few salient points

Meta-atoms/molecules are ‘delicate’ sub wavelength structures containing both the metal & dielectric.

The response of the (composite) meta-atoms of meta material to an e.m. field is ‘critically’ contingent on the specific architecture of meta-atoms, and thus it is difficult to develop a simple and unified method for analyzing the properties of an arbitrary structural unit of meta-material.

Nevertheless, for meta-materials without well structured building blocks there could be generalized analytical methods; that shall enable us to estimate the average e.m. behavior & response of composite materials.

In such randomly structured media with metal and dielectric components are arranged in a disordered fashion, and the overall optical properties of metal-dielectric composites can be significantly different from those of its constituent materials. Very direct consequence we see in cooking a dish which has a overall different taste significantly different from the tastes of individual ingredients.

In a composite where metal-dielectric composites intermingles in a ‘disordered’ manner, the boundary conditions in the system are very complicated that solving Maxwell equation to determine the overall e.m response is rather impossible.

Fortunately one can simplify this disordered system under certain conditions

About mixing rules-effective medium theory

For the study of optical behavior (properties) of these composites with in-homogeneity scale much smaller than the wavelength of excitation signal, electrodynamic scattering by individual metal or dielectric particles is overtaken by overall average response of the entire system.

Therefore we can study the optical properties of these microscopically heterogeneous systems by evaluating the effective dielectric function considering the system to be macroscopically homogeneous.

Thus we can obtain the 'effective dielectric' function in terms of the permittivity of the individual constituents and their mixing ratios in terms of volume fractions.

This is called 'Effective Medium Theory'.

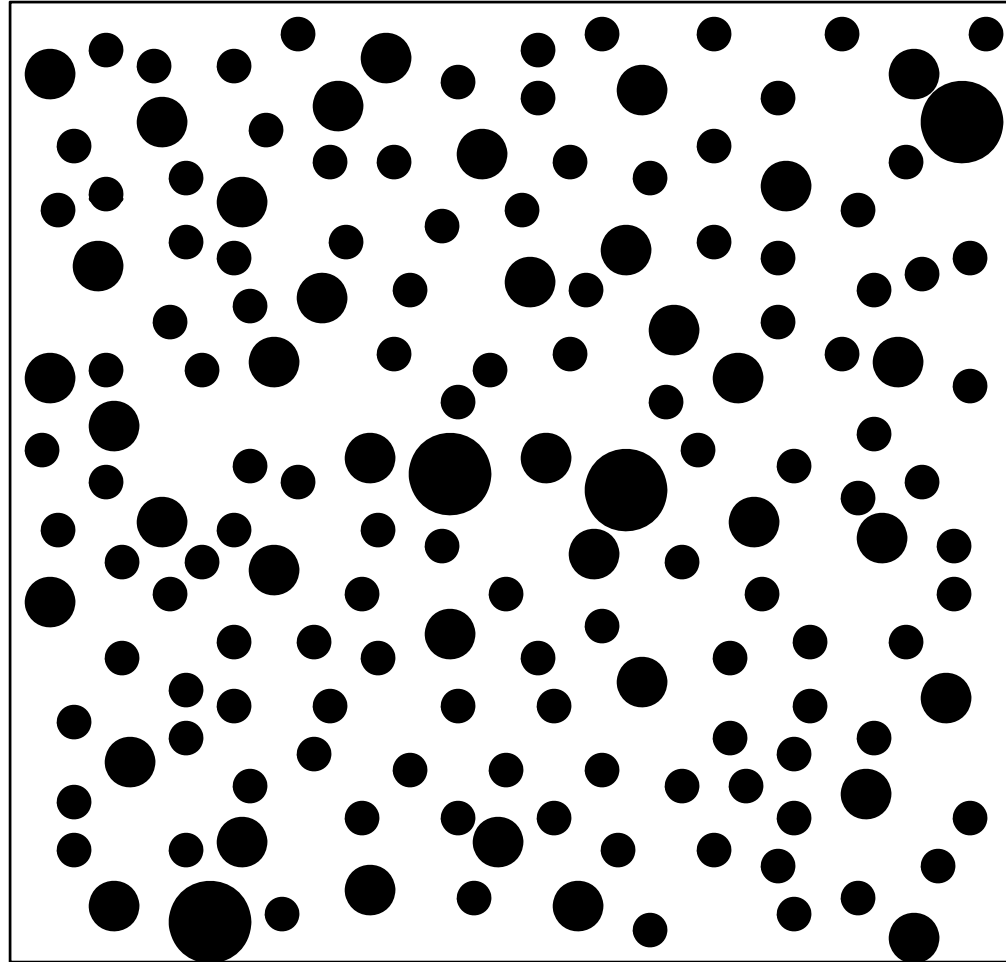
The two theories used are (i) The Maxwell Garnett Theory (MGT) and (ii) Bruggeman Effective Medium Theory (EMT). These two theories distinguish by the microscopic topology used.

The effective medium approximation is also called mean field theory apart from basic MGT and EMT the modern approach is via Bergman-Milton Theory to use spectral density function (very complicated approach)

We will elaborate and restrict to MGT and EMT to get to feel the 'effective medium' behavior of composites

Maxwell-Garnett Geometry

The inclusions are embedded into host



A typical MGT geometry from TEM-image of composite.

Black: are metal, White : are dielectric; the scale bar is 200 nm.

What does Maxwell-Garnet Geometry say?

TEM image of the sample displays MGT topology, the inclusions metal (black); is embedded in Host dielectric (white).

The inclusions (black) are quite dilute.

The inclusions have spherical shape.

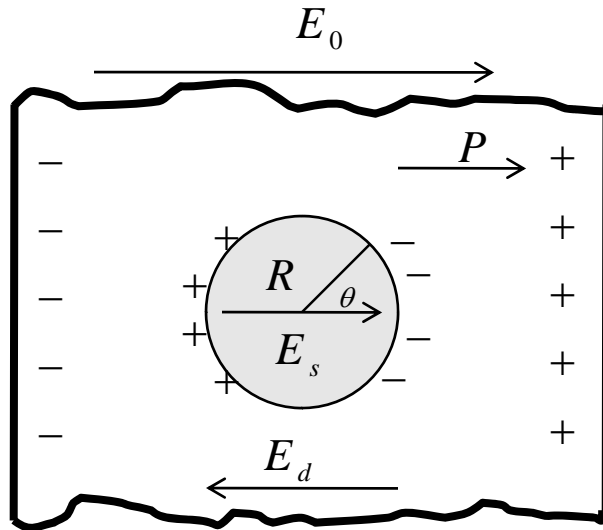
The ‘Local’ field (Lorentz)-and to evaluate the local field

Without loss of generality, consider a dense optical medium. With molecular dipoles arranged in cubic lattice. It was pointed out first by Lorentz, that the local field experienced by a molecule, is ‘not’ the macroscopically averaged field E , but a local field called E_L (L implies local) . This is fact as there are indeed giant fields inside an atom or within the gaps between atoms in solids, because all solids are shown to be non-uniform when examined at atomic scale. However, all these local fluctuations are averaged out to zero if you look at the material at a much larger scale than that of atomic features.

Macroscopically thus, the magnitude of the field in a homogeneous medium is regarded as constant if loss is not an issue. However, when we study the effect of an external field upon an individual atom or molecule or particle, the local features of e.m. fields must be carefully analyzed.

To evaluate the local field E_L at the site of say molecule in a uniform solid, the molecule is ‘imagined’ to be surrounded by a spherical cavity (called Lorentz sphere). The radius R of the sphere is macroscopically small in order to accommodate the discrete nature of the medium very close to the molecule, but it is microscopically large enough so that the matrix just outside be treated as continuous medium. The space inside the sphere is free space with dielectric constant ϵ_0 , that of vacuum, because the gap between the individual molecules contains nothing but free-space. When the external electric field is applied, electric charges are distributed around the surface of Lorentz sphere, which gives rise to additional field imposed upon the central molecule.

The Lorentz sphere it's surface charge distribution and local field



The local field acting on the central dipole, can be decomposed as sum of four components:

$$E_L = E_0 + E_d + E_s + E_{near}$$

E_0 :The external field

E_d :The depolarization field, is due to the polarization charges lying at external surface of medium

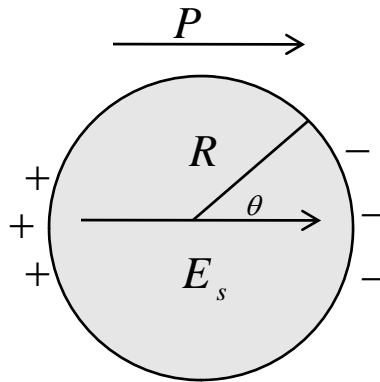
P : The macroscopic (volume) polarization is equal related to depolarization field as $E_d = -P / \epsilon_0$

E_s : Denotes the field due to polarization charges lying on surface of Lorentz sphere

E_{near} :The field induced by other charges lying within the sphere

The term $E_0 + E_d$ in the local field expression, is the homogeneous field averaged over the entire volume, it is exactly the macroscopic field E that enters the Maxwell's equation. For a homogeneous medium this E has a constant magnitude through out the medium. On the other hand the local field E_L is the microscopic field that fluctuates rapidly within the medium, and that can be giant like, at the molecular sites.

Simple calculations for term E_s in local field expression

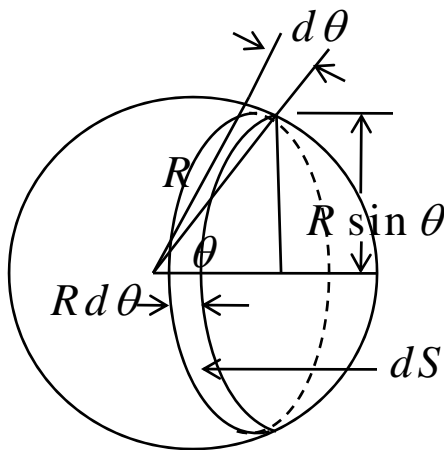


$$E_L = E_0 + \underbrace{E_d}_{=E} + E_s + E_{near}$$

Relate surface charge density on the surface of Lorentz sphere to the polarization P in the medium. That is total charge over a surface segment dS is $(P \cos \theta) dS$; where θ is the angle between P and the normal of the surface segment. This amount of surface charge produces an electric field dE_s , at a distance R (in radial direction), given by:

$$dE_s = \frac{(P \cos \theta) dS}{4\pi\epsilon_0 R^2}$$

The total field E_s resulting from all the surface charges on the Lorentz sphere is directed along the external field E_0 , with a magnitude (horizontal component)



$$E_s = \int_s \frac{(P \cos \theta) dS}{4\pi\epsilon_0 R^2} \cos \theta = \int_s \frac{(P \cos^2 \theta)}{4\pi\epsilon_0 R^2} dS$$

The circular strip is dS having circumference as $2\pi(R \sin \theta)$
Thus area of strip is $dS = (2\pi R \sin \theta)(R d\theta)$

$$E_s = \int_s \frac{(P \cos^2 \theta)}{4\pi\epsilon_0 R^2} 2\pi R^2 \sin \theta d\theta$$

The calculations for E_s and then for total Lorentz field

$$E_s = \int_s \frac{(P \cos^2 \theta)}{4\pi\epsilon_0 R^2} 2\pi R^2 \sin \theta d\theta = \int_{\theta=0}^{\theta=\pi} \frac{(P \cos^2 \theta)}{4\pi\epsilon_0 R^2} 2\pi R^2 \sin \theta d\theta$$

$$= \frac{P}{2\epsilon_0} \int_0^\pi \cos^2 \theta \sin \theta d\theta$$

$$u = \cos \theta; \quad du = -\sin \theta d\theta \quad E_s = -\frac{P}{2\epsilon_0} \int_1^{-1} u^2 du$$

$$E_s = -\frac{P}{2\epsilon_0} \left[\frac{u^3}{3} \right]_1^{-1} = -\left(\frac{P}{2\epsilon_0} \right) \left[\frac{-1}{3} - \frac{1}{3} \right] = \frac{P}{3\epsilon_0}$$

$$E_L = E_0 + E_d + E_s + E_{near} = E + E_s = E + \frac{P}{3\epsilon_0}; \quad E_{near} = 0$$

We assumed E_{near} , the field due to dipoles within the spherical cavity to be zero. Actually this field depends upon the crystal structure, in liquids or gases it vanishes, where the dipoles are randomly distributed in uncorrelated positions. In this example of solid in our case, we assumed a cubic crystal lattice, where E_{near} vanishes owing to the lattice symmetry. The expression for total local Lorentz field is

$$E_L = E + \frac{P}{3\epsilon_0}$$

The field acting at an atom/molecule/particle site is macroscopic field E plus $P / 3\epsilon_0$, from polarization of other atoms/molecules/particles in the system: This is Lorentz relation

Polarizability and Clausius-Mossotti relation (macroscopic property related to microscopic parameter)

We relate polarization P to the electric dipole moment of each atom/molecule/particle; like done in Drude models. Denote α as polarizability of one atom/molecule/particle; if N denotes volume density of dipoles then:

$$P = N \alpha E_L = N \alpha \left(E + \frac{P}{3 \epsilon_0} \right) \quad \text{gives} \quad \frac{N \alpha}{3 \epsilon_0} P = P - N \alpha E$$

We also have constitutive relation, in frequency domain as: $D = \epsilon_0 E + P = \epsilon_0 (1 + \chi_e) E = \epsilon_0 \epsilon_r E$

Use from this (dropping subscript r) $P = \epsilon_0 (\epsilon - 1) E$ in above and via following steps we obtain

$$\frac{N \alpha}{3 \epsilon_0} P = P - N \alpha E$$

$$\frac{N \alpha}{3 \epsilon_0} \epsilon_0 (\epsilon - 1) E = \epsilon_0 (\epsilon - 1) E - N \alpha E$$

$$\frac{N \alpha}{3} (\epsilon - 1) + N \alpha = \epsilon_0 (\epsilon - 1)$$

$$\frac{N \alpha}{3} [(\epsilon - 1) + 3] = \epsilon_0 (\epsilon - 1)$$

$$\frac{N \alpha}{3 \epsilon_0} = \frac{\epsilon - 1}{\epsilon + 2}$$

$$\text{Polarizability } \alpha = \frac{3 \epsilon_0}{N} \frac{(\epsilon - 1)}{(\epsilon + 2)}$$

This is Clausius-Mossotti relation, which gives the necessary link between the macroscopic observable ϵ and the microscopic parameter α .

Application of Clausius-Mossotti relation

The Clausius-Mossotti relation relating the macroscopic observables to microscopic parameter is not mysterious, because there is a distinct connection between the electric response of individual molecule and the macroscopic behavior of the bulk material described by dielectric function (susceptibility).

Now we apply this obtained relation to metal-dielectric composites.

We assume the spherical particles of relative dielectric permittivity ϵ_1 are embedded in the host medium of relative permittivity as ϵ_h ; and we re-write the Clausius-Mossotti relation as:

$$\frac{N\alpha}{3\epsilon_0} = \frac{\epsilon - 1}{\epsilon + 2} \quad \text{becomes} \quad \frac{N\alpha}{3\epsilon_0\epsilon_h} = \frac{\epsilon - \epsilon_h}{\epsilon + 2\epsilon_h} \quad \text{or} \quad \alpha = \frac{3\epsilon_0\epsilon_h}{N} \left(\frac{\epsilon - \epsilon_h}{\epsilon + \epsilon_h} \right)$$

In above ϵ represents the effective permittivity of the composite. Note that $(1 / N)$ is volume occupied by each molecule; in above relation.

If f is the filling fraction of the material in the host i.e. with ϵ_1 . Then we can rewrite above as

$$\alpha = \frac{3\epsilon_0\epsilon_h f}{N} \left(\frac{\epsilon_1 - \epsilon_h}{\epsilon_1 + \epsilon_h} \right)$$

From above we write effective permittivity as

$$\frac{\epsilon - \epsilon_h}{\epsilon + 2\epsilon_h} = f \frac{\epsilon_1 - \epsilon_h}{\epsilon_1 + 2\epsilon_h}$$

Effective Medium (Maxwell-Garnet Theory MGT)

$$\frac{\varepsilon - \varepsilon_h}{\varepsilon + 2\varepsilon_h} = f \frac{\varepsilon_1 - \varepsilon_h}{\varepsilon_1 + 2\varepsilon_h} = fx \quad x = \frac{\varepsilon_1 - \varepsilon_h}{\varepsilon_1 + 2\varepsilon_h}$$
$$\varepsilon - \varepsilon_h = \varepsilon fx + 2\varepsilon_h fx \quad \varepsilon(1 - fx) = \varepsilon_h(1 + 2fx)$$
$$\varepsilon = \varepsilon_h \frac{1 + 2fx}{1 - fx} = \varepsilon_h \frac{1 + 2f \frac{\varepsilon_1 - \varepsilon_h}{\varepsilon_1 + 2\varepsilon_h}}{1 - f \frac{\varepsilon_1 - \varepsilon_h}{\varepsilon_1 + 2\varepsilon_h}}$$

This is known as Maxwell-Garnet formula, describing the ‘bulk’ effective permittivity of a composite in terms of the permittivity of inclusion ε_1 and the host dielectric ε_h . For metal-dielectric, we can view the metal as the inclusion while the dielectric component serves as host.

Although the effective permittivity in MGT can reach the permittivity of the two constituents when the f approaches the two extremes $f=0$ and $f=1$, giving $\varepsilon = \varepsilon_h$; $\varepsilon = \varepsilon_1$, the MGT formula shows that MGT treats the matrix and the inclusion in unsymmetrical fashion. Therefore before evaluating the ε of the two-phase composites, one component should be treated as host and the other one as inclusion. This asymmetry is particularly strong when the difference in the permittivity of the two are large. In fact the MGT gives reasonable estimation of effective ε only when the volume filling fraction f of the inclusion is very small than unity

Approximation to Maxwell-Garnet formula

$$x = \frac{\varepsilon_1 - \varepsilon_h}{\varepsilon_1 + 2\varepsilon_h} \quad \varepsilon = \varepsilon_h \frac{1 + 2fx}{1 - fx} \quad f \ll 1$$

$$\varepsilon = \varepsilon_h (1 + 2fx)(1 - fx)^{-1} \cong \varepsilon_h (1 + 2fx)(1 + fx) = \varepsilon_h [1 + 3fx + \mathcal{O}(f^2 x^2)]$$

$$\varepsilon = \varepsilon_h + 3f \varepsilon_h \frac{\varepsilon_1 - \varepsilon_h}{\varepsilon_1 + 2\varepsilon_h} + \mathcal{O}(f^2)$$

Therefore, the MGT formula first order approximation is linear function of f , and does not yield a critical threshold f_c for the metal-insulator phase transition of a composite.

The resonance is occurring at $\varepsilon_1 = -2\varepsilon_h$ which represents surface plasmon resonance of an isolated metal spherical inclusion embedded in the host dielectric.

From the Drude model for noble metal $\varepsilon_1 = \varepsilon_m = 1 - \frac{\omega_p^2}{\omega^2} = -2\varepsilon_h$; gives $\omega = \omega_p / \sqrt{1 + 2\varepsilon_h}$ as the surface plasmon resonance frequency.

Plot of absorption of dilute metal suspension in dielectric vacuum $\epsilon_h = 1$

We defined absorption in transparent medium as $\alpha \triangleq 4\pi n'' / \lambda_0$, where n'' is imaginary part of refractive index $n = n' + in'' = \sqrt{\epsilon}$. We can write $n'' = \text{Im} \sqrt{\epsilon}$ and absorption as

$$\alpha = 4\pi n'' / \lambda = 2(2\pi / \lambda) \text{Im} \sqrt{\epsilon} = (2\omega / c) \text{Im} \sqrt{\epsilon} \quad \epsilon = \epsilon_h + 3f\epsilon_h \frac{\epsilon_1 - \epsilon_h}{\epsilon_1 + 2\epsilon_h} \quad f = 0.05$$

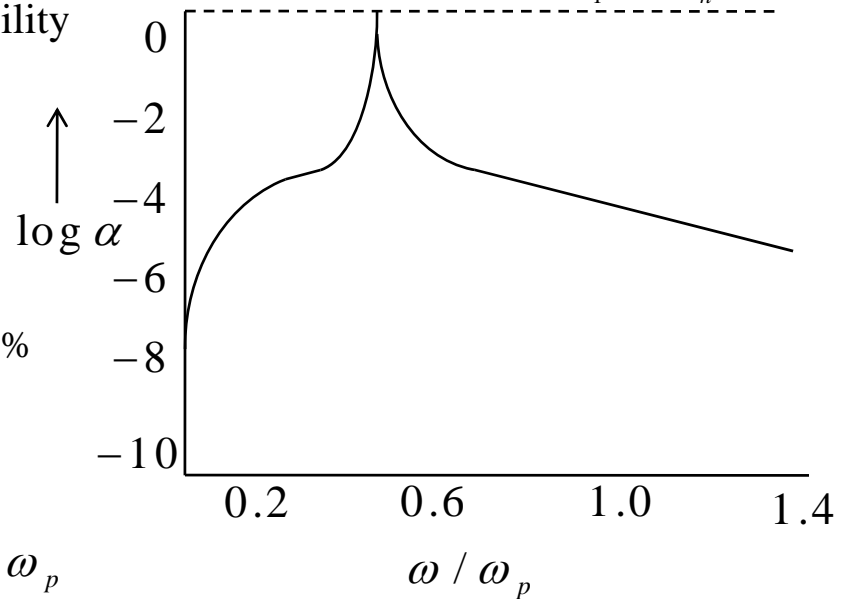
Note: Here α is absorption & not polarizability

$$\begin{aligned} \epsilon_1(\omega) &= 1 - \frac{\omega_p^2}{(\omega^2 + i\omega\Gamma)} \\ &= 1 - \frac{\omega_p^2}{\omega^2 + \Gamma^2} + i \frac{\omega_p^2 \Gamma}{\omega(\omega^2 + \Gamma^2)} \end{aligned}$$

$$\Gamma = 0.002\omega_p \quad \text{Metal filling} \quad f = 0.05 \quad \text{or} \quad 5\%$$

$$\omega = \omega_p / \sqrt{1 + 2\epsilon_h} = \omega_{SPR}$$

$$\epsilon_h = 1 \quad \omega = \omega_p / \sqrt{3} = 0.57\omega_p$$



The spectral position of the prominent absorption peak approaches the surface plasmon resonance Frequency of metal as given by $\omega = \omega_p / \sqrt{1 + 2\epsilon_h}$ in this case $\omega_p / \sqrt{3} \sim 0.57\omega_p$

Why restrict to low filling fraction f ?

In order to evaluate effective medium parameters ε without restriction of low f Bruggeman improved the MGT formula. Bruggeman treated the two parameters ε_1 that of inclusion and ε_h that of host 'with symmetry'.

Therefore there is no such identification as of inclusion and host.

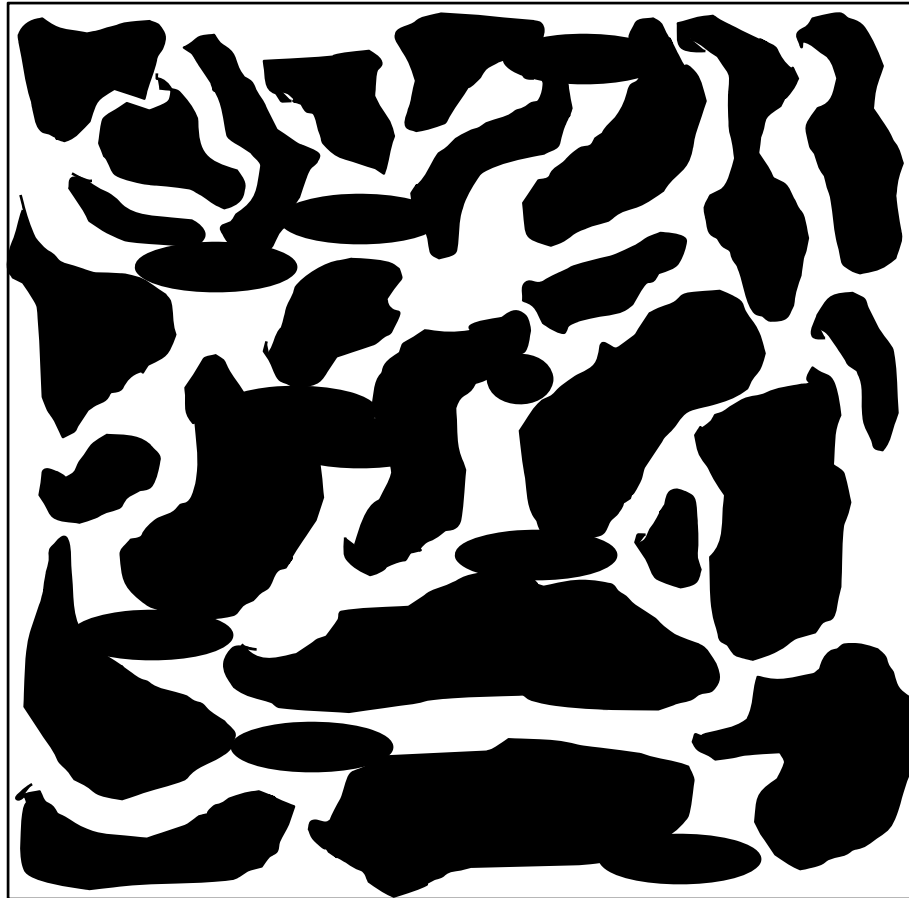
Both are treated equally

Therefore Bruggeman could overcome the limitations of the MGT formulations and the restriction of inclusion having low volume filling fraction

Bruggeman Geometry

The inclusion and host are symmetrical cannot distinguish who is who!!

→ | 200 nm | ←



A typical Bruggeman geometry from TEM-image of composite.

Black: are metal, White : are dielectric; the scale bar is 200 nm.

Bruggeman Effective Medium Theory (EMT)

Let us consider spherical particles of two different materials (ϵ_1, ϵ_2) that are dispersed in a host matrix with ϵ_h . The volume filling fractions of the two inclusions are say (f_1, f_2) . In this case the MGT formula gets modified as

$$\begin{aligned}\frac{\epsilon - \epsilon_h}{\epsilon + 2\epsilon_h} &= \frac{N\alpha}{3\epsilon_0\epsilon_h} & \alpha &= \frac{3\epsilon_0\epsilon_h}{N} \frac{\epsilon_1 - \epsilon_h}{\epsilon_1 + 2\epsilon_h} \\ \frac{\epsilon - \epsilon_h}{\epsilon + 2\epsilon_h} &= \frac{N_1\alpha_1}{3\epsilon_0\epsilon_h} + \frac{N_2\alpha_2}{3\epsilon_0\epsilon_h} \\ &= \frac{N_1}{3\epsilon_0\epsilon_h} \frac{3\epsilon_0\epsilon_h f_1}{N_1} \frac{\epsilon_1 - \epsilon_h}{\epsilon_1 + 2\epsilon_h} + \frac{N_2}{3\epsilon_0\epsilon_h} \frac{3\epsilon_0\epsilon_h f_2}{N_2} \frac{\epsilon_2 - \epsilon_h}{\epsilon_2 + 2\epsilon_h} \\ \frac{\epsilon - \epsilon_h}{\epsilon + 2\epsilon_h} &= f_1 \frac{\epsilon_1 - \epsilon_h}{\epsilon_1 + 2\epsilon_h} + f_2 \frac{\epsilon_2 - \epsilon_h}{\epsilon_2 + 2\epsilon_h}\end{aligned}$$

Clearly the two inclusions are now included in a symmetric manner.

EMT for two phase composite

$$\frac{\varepsilon - \varepsilon_h}{\varepsilon + 2\varepsilon_h} = f_1 \frac{\varepsilon_1 - \varepsilon_h}{\varepsilon_1 + 2\varepsilon_h} + f_2 \frac{\varepsilon_2 - \varepsilon_h}{\varepsilon_2 + 2\varepsilon_h}$$

In a two-phase composite, where $f_1 + f_2 = 1$, each constituent should be regarded as one inclusion, and the 'host' medium is the composite material itself. Therefore in above set $\varepsilon = \varepsilon_h$, to get

$$f_1 \frac{\varepsilon_1 - \varepsilon}{\varepsilon_1 + 2\varepsilon} + f_2 \frac{\varepsilon_2 - \varepsilon}{\varepsilon_2 + 2\varepsilon} = 0 \quad f_1 + f_2 = 1$$

This is quadratic equation and we obtain ε as

$$\varepsilon = \frac{1}{4} \left\{ (3f_1 - 1)\varepsilon_1 + (3f_2 - 1)\varepsilon_2 \pm \sqrt{[(3f_1 - 1)\varepsilon_1 + (3f_2 - 1)\varepsilon_2]^2 + 8\varepsilon_1\varepsilon_2} \right\}$$

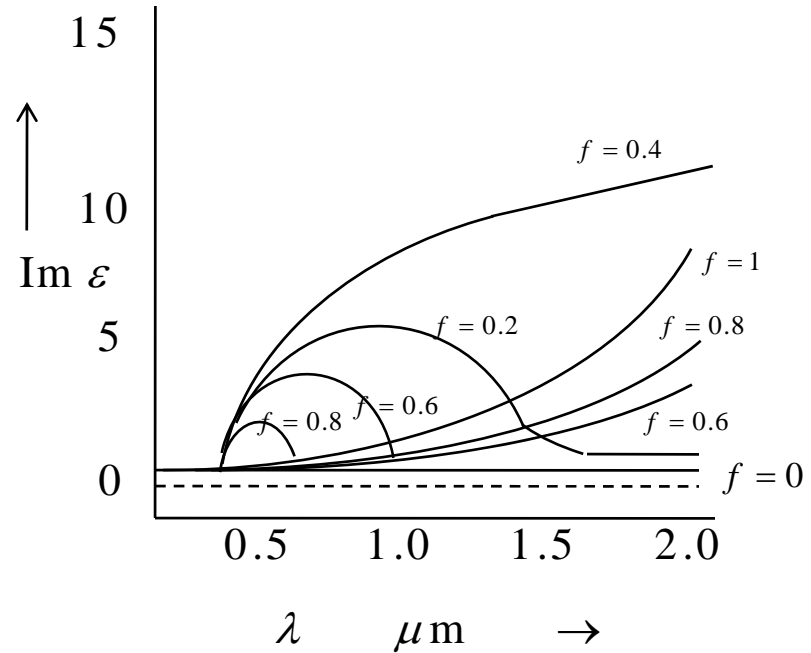
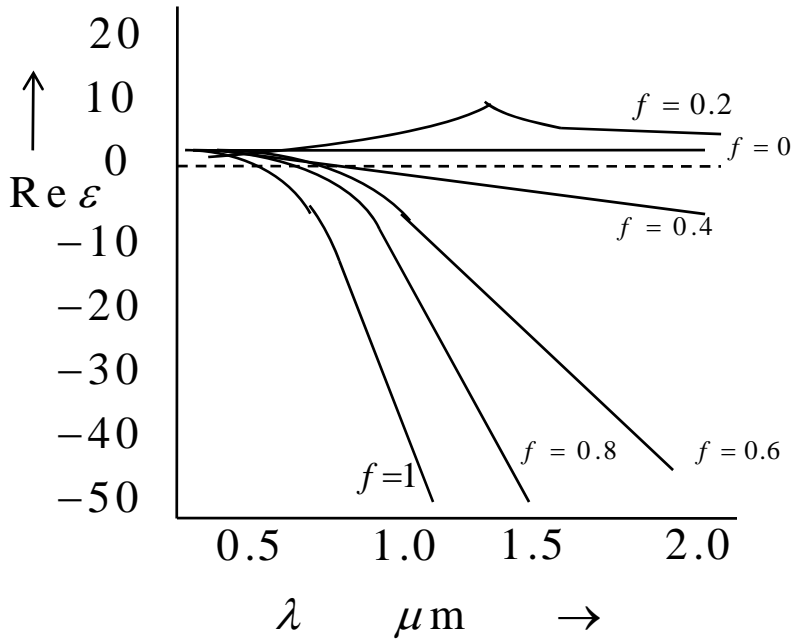
The sign is chosen to make $\text{Im } \varepsilon > 0$; as required by causality in any passive medium

Unlike the MGT the two components here are symmetrical with respect to the exchange roles of $\varepsilon_1; \varepsilon_2$. In EMT thus both the phases are considered to be embedded in the 'effective medium' and there is no preference of one over other. We can thus have for any number of components mixed, as each component in mixture is treated equally.

$$\sum_i f_i \frac{\varepsilon_i - \varepsilon}{\varepsilon_i + 2\varepsilon} = 0 \quad \sum_i f_i = 1$$

Effective permittivity of silver-silica composite for series of metal filling f

$$\varepsilon = \frac{1}{4} \left\{ (3f - 1)\varepsilon_1 + (3[1 - f] - 1)\varepsilon_2 \pm \sqrt{[(3f - 1)\varepsilon_1 + (3[1 - f] - 1)\varepsilon_2]^2 + 8\varepsilon_1\varepsilon_2} \right\}$$



Drude model of silver $\varepsilon_1(\omega) = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + \Gamma^2} + i \frac{\omega_p^2}{\omega(\omega^2 + \Gamma^2)}$; $\varepsilon_\infty = 5$; $\Gamma \approx 0$; $\omega_p = 14 \times 10^{15} \text{ s}^{-1}$

Sellmeir dispersion formula for dielectric silica $\varepsilon_2(\lambda) = n(\lambda)^2 = 1 + \sum_j \frac{S_j \lambda^2}{\lambda^2 - \lambda_j^2}$

Inference from Bergeman (EMT) formula applied for silver-silica composite, the percolation threshold (critical f)

The real part of effective ϵ approaches silver permittivity with increasing f

The imaginary part of ϵ shows a broadening of resonance peak (could be due to e.m. interaction between metal & dielectric grains!)-no concrete explanation on this part of observation.

Apart from the self-consistency symmetry feature another important aspect of EMT predicts critical f . We observe the resonance band for $f = 0.4$ is very broad (in the plot of $\text{Im } \epsilon$). In fact this peak can extend to an infinite bandwidth when f approaches $1/3$. This is percolation threshold for a three-dimensional metal-dielectric composite.

Electronically the percolation threshold represents, the minimum volume fraction f of the conducting particles needed for the formation of a continuous conducting pathway. In a composite where the magnitude of the metal permittivity is much larger than permittivity of dielectric i.e. $|\epsilon_1| \gg \epsilon_2$ then one can approximate the effective permittivity ϵ form

$$\epsilon = \frac{1}{4} \left\{ (3f_1 - 1)\epsilon_1 + (3f_2 - 1)\epsilon_2 \pm \sqrt{[(3f_1 - 1)\epsilon_1 + (3f_2 - 1)\epsilon_2]^2 + 8\epsilon_1\epsilon_2} \right\}$$

as

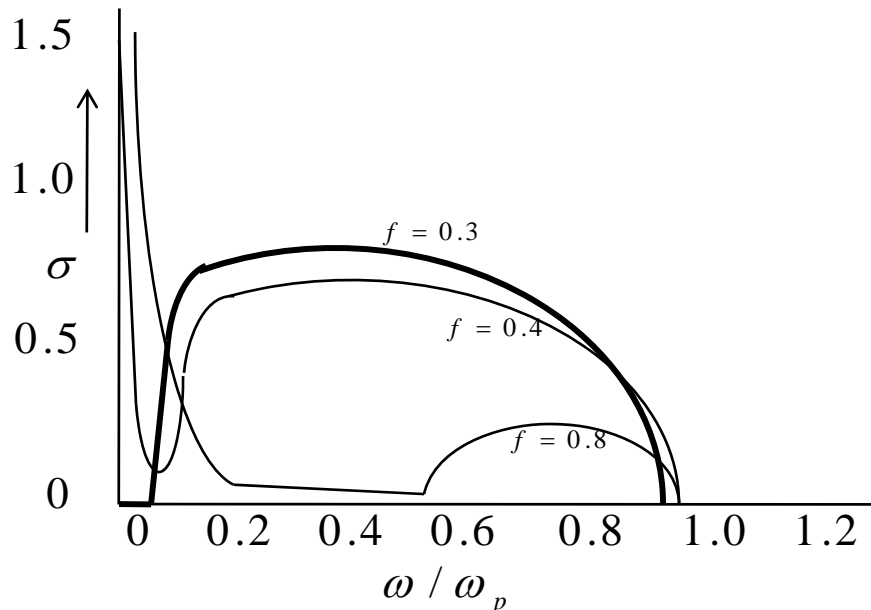
$$\frac{\epsilon}{\epsilon_1} = \begin{cases} \frac{1}{2}(3f_1 - 1); & \text{if } \frac{1}{3} < f_1 \leq 1 \\ 0 & \text{if } 0 < f_1 \leq \frac{1}{3} \end{cases}$$

Discussion about percolation threshold

$$\frac{\varepsilon}{\varepsilon_1} = \begin{cases} \frac{1}{2}(3f_1 - 1); & \text{if } \frac{1}{3} < f_1 \leq 1 \\ 0 & \text{if } 0 < f_1 \leq \frac{1}{3} \end{cases}$$

This clearly indicates that metal-dielectric composite act as a dielectric medium for small metal fraction of less than $1/3$, beyond the percolation threshold the composite acts as dilute metal with an effective permittivity proportional to $[f - (1/3)]$

The sudden change in the electronic-conductivity at the percolation threshold is related to the imaginary part of the effective permittivity as $\sigma(\omega) = \omega[\text{Im } \varepsilon(\omega)]$



Metal-Dielectric (vacuum) conductivity at various f

The interesting feature is that spikes in the σ at low frequency $\omega \rightarrow 0$, when the metal filling f is larger than $1/3$. The broad resonance 'peak' is due to the peaks as in $\text{Im } \varepsilon$ graph.

In other words the composite electronically is like an insulator for dc conductivity when $f < 1/3$, and act as conductor for $f > 1/3$; because a continuous metallic path is formed across the sample and the metal components form an infinite cluster.

Composites in other than three dimensions, the modified MGT and EMT

In the earlier derivations of MGT and then EMT we assumed metal-dielectric composites to be three dimensional; however, there is important category of random media where the composite material is confined to a thin layer with deeply ‘sub wavelength’ thickness-thus making the composite as a two dimensional ‘film’. There is a need to generalize the MGT and EMT for a d -dimensional case. This is done via replacing $(\varepsilon_i + 2\varepsilon)$ by term $[\varepsilon_i + (d - 1)\varepsilon]$; d is the Euclidian dimension of the medium

Generalized MGT is

$$\frac{\varepsilon - \varepsilon_h}{\varepsilon + (d - 1)\varepsilon_h} = f \frac{\varepsilon_1 - \varepsilon_h}{\varepsilon_1 + (d - 1)\varepsilon_h}$$

Generalized EMT is

$$f_1 \frac{\varepsilon_1 - \varepsilon}{\varepsilon_1 + (d - 1)\varepsilon} + f_2 \frac{\varepsilon_2 - \varepsilon}{\varepsilon_2 + (d - 1)\varepsilon} = 0$$

Accordingly the resonance peak in MGT for a low f metal-dielectric film will be $\omega_p / \sqrt{1 + (d - 1)\varepsilon_h}$ and the percolation threshold in EMT will be $f = 1/d$ which is 0.5 for two-dimensional semi-continuous metal film.

The shape factor of the inclusions

In all derivations we assumed inclusion are spherical shape. When the inclusions are notably non-spherical , modification are required in MGT and EMT with additional depolarization or screening factor. To be more general, consider an inclusion of ellipsoidal particles with semi axes $a_i; a_j$ & a_k . In this case the polarizability is as follows:

$$\alpha_{ii} = \frac{\epsilon_0 \epsilon_h}{(\epsilon_1 - \epsilon_h)L_i + \epsilon_h} a_i a_j a_k$$

where L_i is depolarization factor
$$L_i = \int_0^\infty \frac{a_i a_j a_k ds}{2(s + a_i^2)^{3/2} (s + a_j^2)^{1/2} (s + a_k^2)^{1/2}}$$

The depolarization L_i depends on the shape of the particle and the direction of E field of incident light w.r.t. the particle's axis a_i . The values of L_i is between 0 and 1/3 when a particle is prolate along the axis a_i , while the values are between 1/3 and 1 if the particle is disk like. The sum of all three depolarization factors $\sum L_i$, must be equal to unity for any particle shape.

The Numerical integration and graphs for the above are in following references, along with simplified Formulas for special ellipsoids prolate, oblate, spheroids etc.

Osborne JA , (1945) Demagnetizing factors of general ellipsoid. Phys. Rev. 67: 351

Van de Hulst (1981) light scattering by small particles. Dover; New-York.

The MGT and EMT with shape factor depolarization (screening)

The MGT is modified as:
$$\frac{\varepsilon - \varepsilon_h}{\varepsilon + \kappa \varepsilon_h} = f \frac{\varepsilon_1 - \varepsilon_h}{\varepsilon_1 + \kappa \varepsilon_h}$$

The EMT is modified as:
$$f_1 \frac{\varepsilon_1 - \varepsilon}{\varepsilon_1 + \kappa \varepsilon} + f_2 \frac{\varepsilon_2 - \varepsilon}{\varepsilon_2 + \kappa \varepsilon} = 0$$

κ is the screening parameter & is shape and orientation (w.r.t E – field)-dependent of the nano-inclusions. This factor is related to Lorentz depolarization factor L by: $\kappa = (1 - L) / L$

A long ‘elliptical’ cylinder placed along the E field will have Lorentz depolarization factor $L \approx 0$ and thus screening parameter $\kappa \rightarrow \infty$. For an example, is a film of silver wires which gives full screening for light in one polarization but has no screening in the perpendicular direction. This is extreme example, and such films are used as linear polarizer in optical systems.

For spherical nano particles $L = 1/3$ and $\kappa = 2$, thus we get original MGT and EMT

Note that MGT works well when f is small, is reliable only when screening is not substantial. If the L i.e. depolarization is close to zero, there is a strong interaction between nano-particles and the external E field and the system may not behave as shape-dependent MGT even with small f . In this case modified EMT should be used even with small f . Moreover shape sensitive EMT also gives modified percolation threshold as $f_c = L$

Reversed effective medium approximation

These effective medium approximations can be used in reverse, i.e. in nano-crystal research, it is a standard practice to calculate the permittivity of the nano-particles when the permittivity of the other component is known and that of whole component is measured (experimentally).

In such cases Bruggeman EMT is preferred over MGT, because singularities arise in reverse MGT calculations when the f of nano-particles is large, or when contrast in permittivity in two phases is significant.

When non-spherical nano particles are present then modified EMT should be used accordingly.

Conclusions and advancements on ‘mean-field theory’

The discussed effective medium approximations are also called ‘mean field theory’-these are just Analytical approximations to estimate effective permittivity of a composite. Although simple but such Theory fail to address the fine features of sub wavelength sized components.

The most general Effective Medium Approach is the spectral representation method (Bergman Theory or Bergman-Milton representation). Here spectral density functions are used as fit functions and correlate with geometrical details of composite. In the spectral representation theorem the micro- or nano-scale features are no longer simply neglected or described using numbers, like depolarization factor L ‘ but are fully taken into account by defining geometrical functions having general analytical properties. This method clearly distinguishes between the influence of the geometrical structure and that of the dielectric properties of the components on the effective system behavior.

For spherical or nicely aligned ellipsoidal inclusions this method reduces to MGT or EMT.

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End of part-3