# 4

# EFFECTIVE PERMEABILITY OF MIXTURES

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## 4.1 Introduction

The dielectric and magnetic properties of composite media are of importance in many applications where these materials are to be treated macroscopically homogeneous. The concept of effective permittivity is much used to describe the properties of dielectric mixtures in this large-scale regime. Analogously, the effective permeability quantity can be introduced to treat magnetic mixtures, for example composite materials with ferrites or other magnetically non-neutral components. Effective permeability carries information about the average magnetic polarization in the heterogeneous medium, being the ratio between the average magnetic displacement and the applied magnetic field.

It is well-known that the partial differential equations for electrostatic and magnetostatic problems are formally the same: Laplace equation for the potential. Also, the boundary conditions of the cor-

responding quantities are the same. Hence, these two problems are dual. It means that if the other problem has been solved, the result can be used to find out the solution to the dual problem straightforwardly: changing the quantities to their dual counterparts. Because the effective permittivity is a solution to the electrostatic problem, and numerous mixing formulas for this quantity have been presented for dielectric mixtures, using duality it is easy to start studying the effective permeability for the magnetic mixtures. In physics, there are also other problems that lead to the same Laplace equation with corresponding boundary conditions and are therefore dual to the electrostatic and magnetostatic problems. Among these are, for example, the conductivity problem, both for electrical and heat conduction, and the duality can also be applied to calculating the effective viscosity or the diffusion coefficient of a mixture.

Owing to the duality, the restrictions of the applicability of the effective permittivity are the same for effective permeability. The most important limitation is the quasistatic requirement in calculating the polarization, where averaging has to be made in scales smaller than the spatial variation of the incident electromagnetic field. This means that the wavelength of the applied field has to be larger than the characteristic distance of the inhomogeneities of the heterogeneous medium: this length is the size of the scatterers in case of the mixture being discrete (inclusions in a background medium), or the correlation length for the case of continuous random material. Therefore, effective permeability is a low-frequency concept. At higher frequencies, scattering effects take over, and the term effective permeability loses its meaning. The limit for the size of the scatterers is [1]

$$d_{\max} \simeq \frac{\lambda}{2\pi} \tag{1}$$

where $d_{\max}$ is the maximum diameter of the inclusion and $\lambda$ is the wavelength of the incident field.

Although the electrostatic and magnetostatic problems are dual on the level of partial differential equations and boundary conditions, the resulting theoretical quantities, permittivity and permeability are ultimately of secondary nature: the primary physical polarization and loss mechanisms are "hidden" in these, and the comprised interactions are normally different for the dielectric and magnetic cases. For example, characteristic of magnetic mixtures is the nonlinearity of the materials

and the mixture itself. Nonlinearity also entails losses that are normally not encountered in dielectric materials. Another difference between the dielectric and magnetic mixtures is the dependence on the crucial parameter in the magnetic case, the temperature: above Curie temperature, ferromagnetic and ferrimagnetic materials lose their spontaneous magnetization and become paramagnetic.

## 4.2 The Effective Permeability

### *a. The Duality*

The electrostatic and magnetostatic problems are dual for a given geometry; i.e. the unknown functions obey the same partial differential equations and boundary conditions. In source-free regions, both the electric flux density (or displacement) $\overline{D}$ and the magnetic flux density $\overline{B}$ are divergenceless:

$$\nabla \cdot \overline{D} = 0 \tag{2}$$

$$\nabla \cdot \overline{B} = 0 \tag{3}$$

These flux functions are connected to the field functions $\overline{E}$ (the electric field) and $\overline{H}$ (the magnetic field) by the constitutive relations

$$\overline{D} = \epsilon \overline{E} \tag{4}$$

$$\overline{B} = \mu \overline{H} \tag{5}$$

where $\epsilon$ is the permittivity and $\mu$ is the permeability of the material. On boundaries of different dielectric (and different magnetic, respectively) materials with no sources, the field and flux functions have to satisfy the following boundary conditions: across a boundary, the tangential component of the electric and magnetic field and the normal component of the electric and magnetic flux density have to be continuous, due to the Maxwell equations; see, for example, [2].

In solving the electrostatic problem, a potential $\Phi$ is introduced that gives the electric field as

$$\overline{E} = -\nabla \Phi \tag{6}$$

Therefore $\Phi$ obeys the Laplace equation in dielectrically homogeneous source-free regions. For the magnetic problem, a magnetic potential function can be similarly defined and also obeys the Laplace equation.

Consider a region with a partially homogeneous permittivity function; i.e. the volume consists of subregions of different permittivities. Define the effective permittivity of this heterogeneous material as the ratio between the average displacement $< \overline{D} >$ and the average electric field $< \overline{E} >$:

$$< \overline{D} >= \epsilon_{\text{eff}} < \overline{E} > \tag{7}$$

The formula above is also applicable to enumerating the effective permeability $\mu_{\text{eff}}$ of the magnetostatic problem with the same geometry. This is due to duality; the same partial differential equation and the same boundary conditions are satisfied, and therefore the solutions are the same.

### *b. Mixture with Spherical Inclusions*

Consider a magnetically heterogeneous material consisting of a background medium with permeability $\mu_0$ (not necessarily the vacuum permeability), and spherical inclusions of permeability $\mu_1$. This corresponds to two-phase mixture. In low-frequency regime, where the dimensions of the inclusions are smaller than the wavelength, the following quasistatic analysis is valid. Scattering effects from the inclusions are neglected.

Define the effective magnetic permeability $\mu_{\text{eff}}$ as dual of (7):

$$< \overline{B} >= \mu_{\text{eff}} < \overline{H} > \tag{8}$$

The displacement depends on the average magnetic polarization $\overline{J}$ in the medium (the notation of Jones [3] is here followed, where the magnetic polarization is defined corresponding to the polarization $\overline{P}$ in the dielectric problem, and not as the magnetization normally denoted by $\overline{M}$: these two quantities differ in ratio by the free space permeability)

$$< \overline{B} >= \mu_0 < \overline{H} > + < \overline{J} > \tag{9}$$

The average polarization, or the magnetic dipole moment density, is

$$< \overline{J} >= n\overline{j} \tag{10}$$

where $n$ is the number of inclusions per unit volume, and $\overline{j}$ is the magnetic dipole moment of a single inclusion. The magnetic dipole moment depends on the magnetic polarizability of the inclusion $\beta$ and the exciting field $\overline{H}^e$:

$$\overline{j} = \beta \overline{H}^e \tag{11}$$

The exciting field is not the same as the average field but rather similar to the local field, a quantity used in studying polarization phenomena in the microscopic scale (see, for example, [4,5]). It depends on the average polarization surrounding the inclusion and on the shape of the inclusion. For spherical scatterers it is

$$\overline{H}^e = <\overline{H}> + \frac{<\overline{J}>}{3\mu_0} \tag{12}$$

From these equations, the effective permeability can be written as

$$\frac{\mu_{\text{eff}} - \mu_0}{\mu_{\text{eff}} + 2\mu_0} = \frac{n\beta}{3\mu_0} \tag{13}$$

which is the magnetic form of the famous Lorentz-Lorenz formula (also known as the Clausius-Mossotti formula) [6].

The polarizability of a scatterer can be solved basically with two approaches: integrating the product of the susceptibility and the field over the volume, or considering the perturbation on the outside field due to the dipole moment of the scatterer and finding its relation to the average field [7]. The magnetic polarizability of a homogeneous spherical inclusion is the product of the magnetic susceptibility of the inclusion $\mu_1 - \mu_0$, the field ratio inside and outside the inclusion, and its volume $v$ (for calculating the field ratio in the electrostatic case, see for example [8]):

$$\beta = v(\mu_1 - \mu_0)\frac{3\mu_0}{\mu_1 + 2\mu_0} \tag{14}$$

Combining (13) and (14), we get the magnetic Rayleigh mixing formula

$$\frac{\mu_{\text{eff}} - \mu_0}{\mu_{\text{eff}} + 2\mu_0} = f\frac{\mu_1 - \mu_0}{\mu_1 + 2\mu_0} \tag{15}$$

where $f = nv$ is the volume fraction of the inclusion phase in the mixture. In the form of Maxwell-Garnett formula it is [9]

$$\mu_{\text{eff}} = \mu_0 + 3\mu_0 \frac{f\dfrac{\mu_1 - \mu_0}{\mu_1 + 2\mu_0}}{1 - f\dfrac{\mu_1 - \mu_0}{\mu_1 + 2\mu_0}} \tag{16}$$

### c. *Mixture with Ellipsoidal Inclusions*

Consider next the case of ellipsoidal inclusions. The exciting field for an inclusion is (see [1])

$$\overline{H}^e = <\overline{H}> + N_i \frac{<\overline{J}>}{\mu_0} \tag{17}$$

where $N_i$ is the depolarization factor of the ellipsoid that is different in the three axial directions, and $<\overline{J}>$ varies with direction for anisotropic mixtures. Hence the exciting field also depends on the direction. The depolarization factor for an ellipsoid is [10, 11]

$$N_a = \frac{abc}{2} \int_0^\infty \frac{ds}{(s+a^2)\sqrt{(s+a^2)(s+b^2)(s+c^2)}} \tag{18}$$

Here $a, b$, and $c$ are the semiaxes of the ellipsoid and $N_a$ is the depolarization factor in the direction of the $a$-axis. For depolarization factors $N_b$ and $N_c$, interchange $b$ and $a$, and $c$ and $a$ in (18), respectively.

The depolarization factors satisfy

$$N_a + N_b + N_c = 1 \tag{19}$$

for any ellipsoid. A sphere has three equal depolarization factors of $1/3$. The other two special cases correspond to a disc (depolarization factors $1, 0, 0$), and a needle ($0, 1/2, 1/2$). For a general ellipsoid with three different axes, the depolarization factors have to be calculated from the integral (18). Osborn and Stoner have tabulated the depolarization factors of a general ellipsoid [12,13]. For spheroids (ellipsoids of revolution), which have two equal axes, closed-form expressions can be evaluated for the integral (18). Prolate spheroids $(a > b = c)$ have

$$N_a = \frac{1-e^2}{2e^3}\left(\ln\frac{1+e}{1-e} - 2e\right) \tag{20}$$

$$N_b = N_c = \frac{1}{2}(1 - N_a) \tag{21}$$

where the eccentricity is $e = \sqrt{1 - b^2/a^2}$. For nearly spherical spheroids, which have small eccentricity, the following holds:

$$N_a \simeq \frac{1}{3} - \frac{2}{15}e^2 \tag{22}$$

$$N_b = N_c \simeq \frac{1}{3} + \frac{1}{15}e^2 \tag{23}$$

On the other hand, for nearly needle-shaped prolate spheroids (large eccentricity):

$$N_a \simeq (b/a)^2 \left(\ln \frac{2}{b/a} - 1\right) \tag{24}$$

$$N_b = N_c \simeq \frac{1}{2}\left[1 - (b/a)^2 \left(\ln \frac{2}{b/a} - 1\right)\right] \tag{25}$$

For oblate spheroids $(a = b > c)$,

$$N_c = \frac{1+e^2}{e^3}(e - \arctan e) \tag{26}$$

$$N_a = N_b = \frac{1}{2}(1 - N_c) \tag{27}$$

where $e = \sqrt{a^2/c^2 - 1}$. For nearly-spherical oblate spheroids,

$$N_c \simeq \frac{1}{3} + \frac{2}{15}e^2 \tag{28}$$

$$N_a = N_b \simeq \frac{1}{3} - \frac{1}{15}e^2 \tag{29}$$

and for nearly disc-shaped spheroids,

$$N_c \simeq 1 - \frac{\pi}{2}(c/a) + 2(c/a)^2 \tag{30}$$

$$N_a = N_b \simeq \frac{\pi}{4}(c/a) - (c/a)^2 \tag{31}$$

The magnetic polarizability of a general ellipsoid with volume $v$ and permeability $\mu_1$ has also three components: in the direction of the $a$ axis, it is

$$\beta_a = v(\mu_1 - \mu_0)\frac{\mu_0}{\mu_0 + N_a(\mu_1 - \mu_0)} \tag{32}$$

For a mixture with $n$ ellipsoids of this type per unit volume, if all the ellipsoids are aligned with corresponding axes oriented in the same direction, the mixture is anisotropic and its effective permeability dyadic, or tensor is

$$\overline{\overline{\mu}}_{\text{eff}} = \mu^a_{\text{eff}}\hat{a}\hat{a} + \mu^b_{\text{eff}}\hat{b}\hat{b} + \mu^c_{\text{eff}}\hat{c}\hat{c} \tag{33}$$

where hat denotes unit vector in the direction of the axis, and the permeability components are

$$\mu_{\text{eff}}^{i} = \mu_0 + \frac{\mu_0 f \dfrac{\mu_1 - \mu_0}{\mu_0 + N_i(\mu_1 - \mu_0)}}{1 - f\dfrac{N_i(\mu_1 - \mu_0)}{\mu_0 + N_i(\mu_1 - \mu_0)}} \tag{34}$$

for $i = a, b,$ and $c$ and $f = nv$.

In the case that the ellipsoids are not aligned, the orthogonal components of the dipole moment of a single scatterer contribute not only to one component of the magnetic polarization. As a result, in calculating the average polarization, the dipole moments of all scatterers have to be integrated over their orientation distribution functions [1]. For randomly oriented inclusions there is no preferred direction in the mixture and the effective permeability is isotropic. Averaging the dipole moment contributions gives the effective permeability (which has also been derived by other approaches [14,15] besides [1])

$$\mu_{\text{eff}} = \mu_0 + \frac{\mu_0 \dfrac{f}{3} \displaystyle\sum_{i=a,b,c} \dfrac{\mu_1 - \mu_0}{\mu_0 + N_i(\mu_1 - \mu_0)}}{1 - \dfrac{f}{3} \displaystyle\sum_{i=a,b,c} \dfrac{N_i(\mu_1 - \mu_0)}{\mu_0 + N_i(\mu_1 - \mu_0)}} \tag{35}$$

For randomly oriented needles,

$$\mu_{\text{eff}} = \mu_0 + f(\mu_1 - \mu_0)\frac{\mu_1 + 5\mu_0}{(3 - 2f)\mu_1 + (3 + 2f)\mu_0} \tag{36}$$

and for randomly oriented discs,

$$\mu_{\text{eff}} = \mu_0 + f(\mu_1 - \mu_0)\frac{2\mu_1 + \mu_0}{(3 - f)\mu_1 + f\mu_0} \tag{37}$$

### *d. Other Mixing Rules*

There is no exact result for the effective permeability of a mixture with random geometry. In the analysis of random media, a major

difficulty lies how to correctly take into account the interaction between the scatterers. For sparse mixtures, these effects of interaction are small and can be included by surrounding the inclusion with the average magnetic polarization $< \overline{J} >$ as was the case in the previous analysis. However, in the dense mixture regime, i.e. high scatterer volume fractions, this approach may not be correct. In the following, mixing rules are presented (that have been originally derived for dielectric mixtures) for dense magnetic mixtures which predict different results compared to the Maxwell-Garnett type formulas discussed in the previous section. For more detailed discussion, see [17] and references therein.

The multitude of rivaling mixing formulas that still are applicable undermines the idea of attempting to derive an expression which is valid for all mixtures. This fact has directed Hashin's and Shtrikman's attention to finding upper and lower limits for the effective permeability. In [16], they exploit stationary principles derivable through variational approach, and give the following limits for mixing formulas:

For a two-phase mixture with spherical inclusions and with the condition $\mu_1 > \mu_0$, the effective permeability is bounded below by

$$\mu_{\text{eff}} \geq \mu_0 + \frac{f}{\frac{1}{\mu_1 - \mu_0} + \frac{1-f}{3\mu_0}} \tag{38}$$

and bounded above by

$$\mu_{\text{eff}} \leq \mu_1 + \frac{1-f}{\frac{1}{\mu_0 - \mu_1} + \frac{f}{3\mu_1}} \tag{39}$$

It is interesting to note that the lower limit is exactly the Maxwell-Garnett permeability (16). This agrees with the observation that of all mixing rules, the Maxwell-Garnett always seems to render the smallest result. In the Hashin-Shtrikman bounds, it is also seen the symmetry: the upper bound is the inverted Maxwell-Garnett formula; i.e. $\mu_1$ is the permeability of the background and that of the inclusion is $\mu_0$. From these bounds, the perturbative limits can be written for small fractional volumes of the inclusion phase ($f \ll 1$) that a tentative mixing formula should satisfy:

$$\mu_0 + 3f\mu_0 \frac{\mu_1 - \mu_0}{\mu_1 + 2\mu_0} \leq \mu_{\text{eff}} < \mu_0 + f(\mu_1 - \mu_0) \tag{40}$$

Hence, these are the sparse-mixture limits.

For randomly oriented ellipsoidal inclusions with permeability $\mu_1$ in the background medium of permeability $\mu_0$, Polder–van Santen mixing formula [17,18,19,20] is

$$\mu_{\text{eff}} = \mu_0 + \frac{f}{3}(\mu_1 - \mu_0) \sum_{i=a,b,c} \frac{\mu_{\text{eff}}}{\mu_{\text{eff}} + N_i(\mu_1 - \mu_{\text{eff}})} \tag{41}$$

where $f$ is the volume fraction of the inclusion phase. For spherical inclusions, this simplifies to the Böttcher mixing formula [21]

$$\frac{\mu_{\text{eff}} - \mu_0}{3\mu_{\text{eff}}} = f\frac{\mu_1 - \mu_0}{\mu_1 + 2\mu_{\text{eff}}} \tag{42}$$

The following is the CP formula [1,17]:

$$\mu_{\text{eff}} = \mu_0 + \frac{f}{3}(\mu_1 - \mu_0) \sum_{i=a,b,c} \frac{(1 + N_i)\mu_{\text{eff}} - N_i\mu_0}{\mu_{\text{eff}} + N_i(\mu_1 - \mu_0)} \tag{43}$$

where "CP" originates from the fact that it equals the low-frequency result of the random-medium analysis with "quasicrystalline approximation and coherent potential" for spherical scatterers [20,22]. (Sometimes it is also referred to as QCA-CP.) The formula for spherical inclusions is

$$\mu_{\text{eff}} = \mu_0 + f(\mu_1 - \mu_0)\frac{3\mu_{\text{eff}}}{3\mu_{\text{eff}} + (1 - f)(\mu_1 - \mu_0)} \tag{44}$$

These two mixing rules are implicit equations for the effective permeability. Although for spherical cases $\mu_{\text{eff}}$ can be solved from a second-order equation, the rules have been seen to be suitable for iterative solution. It is also worth noting that for sparse mixtures, Maxwell-Garnett, Polder–van Santen, and CP, all predict the same results up to the first order expansion in $f$.

For dielectric mixtures with spherical inclusions, three mixing formulas have been derived through differential analysis. These formulas contain one-third powers of the permittivities in the problem. Expressed for permeabilities, they are

- Looyenga formula [23]

$$\mu_{\text{eff}}^{1/3} = f\mu_1^{1/3} + (1 - f)\mu_0^{1/3} \tag{45}$$

- Bruggeman formula [24]

$$\frac{\mu_1 - \mu_{\text{eff}}}{\mu_1 - \mu_0} = (1 - f)\left(\frac{\mu_{\text{eff}}}{\mu_0}\right)^{1/3} \tag{46}$$

- Sen, Scala, and Cohen formula [25]

$$\frac{\mu_{\text{eff}} - \mu_0}{\mu_1 - \mu_0} = f\left(\frac{\mu_{\text{eff}}}{\mu_1}\right)^{1/3} \tag{47}$$

For closer analysis on these mixing rules, see [17] and [26].

There are also formulas for mixtures with spherical inclusions in a cubic array in a background matrix. These formulas can be seen as successive improvements to the classical Rayleigh result, (15), [27]. These have been presented by Runge [28], Meredith and Tobias [29], McPhedran, McKenzie and Derrick [30,31], Doyle [32], and by Lam [33]. However, these formulas are derived for ordered mixtures, though not all necessarily for cubic centered lattices, and from the point of view of application to random media, they suffer from the disadvantage of predicting infinite effective permeabilities as the inclusions come to contact with each other.

### *e. Illustrations*

Mixing rules presented in previous sections for the effective permeability of a heterogeneous material depend on the permeabilities of the constituents, their volume fractions, and the shape of the inclusions. To illustrate the effect of these parameters, Figs. 4.1 – 4.5 depict the normalized effective permeability. The abscissa is the volume fraction of the inclusion phase $f$ and the ordinate is the effective magnetic susceptibility $\mu_{\text{eff}} - \mu_0$ normalized by the inclusion phase susceptibility $\mu_1 - \mu_0$. Here, the term susceptibility is defined relative to the background permeability $\mu_0$. The different curves in the figures are for different permeability values of the inclusion phase. Figures 4.1 – 4.3 show the generalized Maxwell-Garnett formula for spherical (16), needle-shaped (36), and disc-shaped (37) inclusion particles with random orientation, and Figs. 4.4 and 4.5 show the Polder–van Santen formula (42) and the CP formula (44), respectively.

The effect of increasing inclusion-background permeability contrast is clearly seen from the figures. As the contrast increases, the

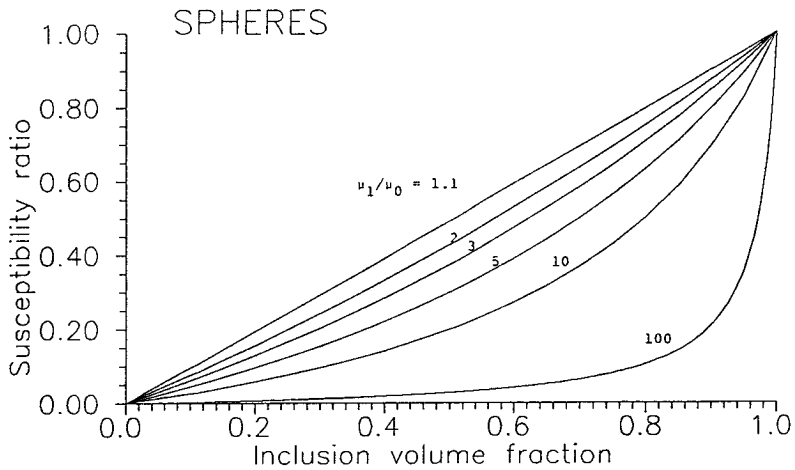


**Figure 4.1 Normalized effective permeability (susceptibility ratio $\mu_{\text{eff}} - \mu_0/\mu_1 - \mu_0$) of a two-phase mixture as a function of the volume fraction of the inclusion phase. The relative inclusion permeability is a parameter. Maxwell-Garnett formula with spherical inclusions.**

effective permeability does not increase in the same proportion for a given volume fraction value. Naturally for the volume fraction value $f = 1$, the effective permeability equals the inclusion permeability, which is the requirement for a consistent mixing formula. The effect of predicting surprisingly low effective permeability values with large inclusion permeabilities is pronounced for the case of spherical inclusions and the Maxwell-Garnett type formula; the Polder–van Santen and CP give higher values. Needles and especially discs predict higher relative values for large inclusion-background permeability contrast, which can be attributed to the fact that the flat surfaces in one (needle) or two (disc) dimensions leads to larger inside fields compared to sphere, hence giving larger dipole moment components in these directions. Properties of mixtures with oblate or prolate spheroidal inclusions fall between the corresponding curves of spherical and disc-shaped or needle-shaped particles.

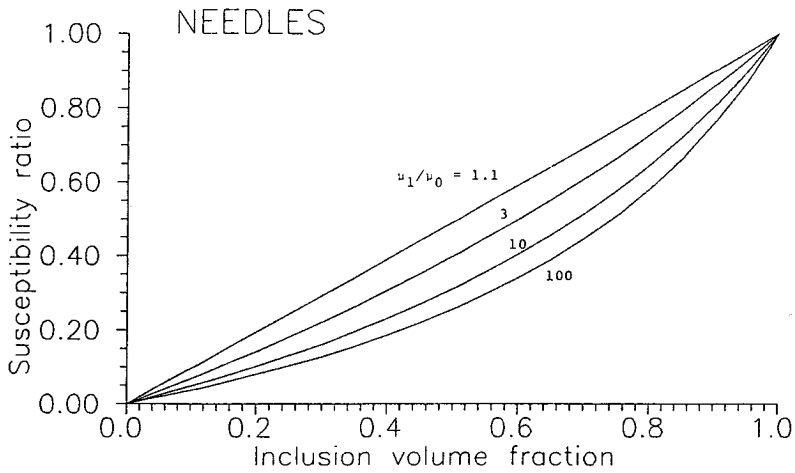


**Figure 4.2 Normalized effective permeability (susceptibility ratio $\mu_{\text{eff}} - \mu_0/\mu_1 - \mu_0$) of a two-phase mixture as a function of the volume fraction of the inclusion phase. The relative inclusion permeability is a parameter. Maxwell-Garnett formula with randomly oriented needle-shaped inclusions.**

## 4.3 Multiphase Mixtures

The previous section discussed two-phase magnetic mixtures, consisting only of one inclusion phase in a homogeneous background medium. However, the analysis is also applicable to multiphase mixtures: media where inclusions of magnetically different species are immersed in a background material, and even, with some limitations to partially and continuously inhomogeneous inclusions. These types of mixtures are treated in this section.

Consider first a $N+1$ –phase mixture with $N$ magnetically different inclusion phases in the background medium with permeability $\mu_0$. Let the inclusions be in the form of spheres of permeabilities $\mu_i$, ($i = 1 \cdots N$), and let the volume fraction of the $i$th phase be $f_i$. The

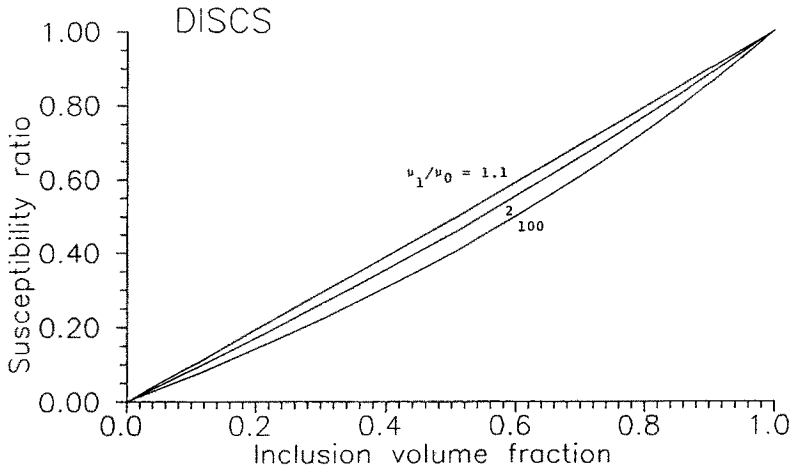


**Figure 4.3 Normalized effective permeability (susceptibility ratio $\mu_{eff} - \mu_0/\mu_1 - \mu_0$) of a two-phase mixture as a function of the volume fraction of the inclusion phase. The relative inclusion permeability is a parameter. Maxwell-Garnett formula with randomly oriented disc-shaped inclusions.**

exciting field for each inclusion is, as in the two-phase case

$$\overline{H}^e = <\overline{H}> + \frac{<\overline{J}>}{3\mu_0} \tag{48}$$

and now the average magnetic polarization is

$$<\overline{J}> = \sum_{i=1}^{N} n_i \overline{j}_i = \sum_{i=1}^{N} n_i \beta_i \overline{H}^e \tag{49}$$

where $\overline{j}_i$ is the magnetic dipole moment of the inclusion with permeability $\mu_i$ and $\beta_i$ its magnetic polarizability. Hence, the effective permeability is

$$\frac{\mu_{eff} - \mu_0}{\mu_{eff} + 2\mu_0} = \sum_{i=1}^{N} \frac{n_i \beta_i}{3\mu_0} \tag{50}$$

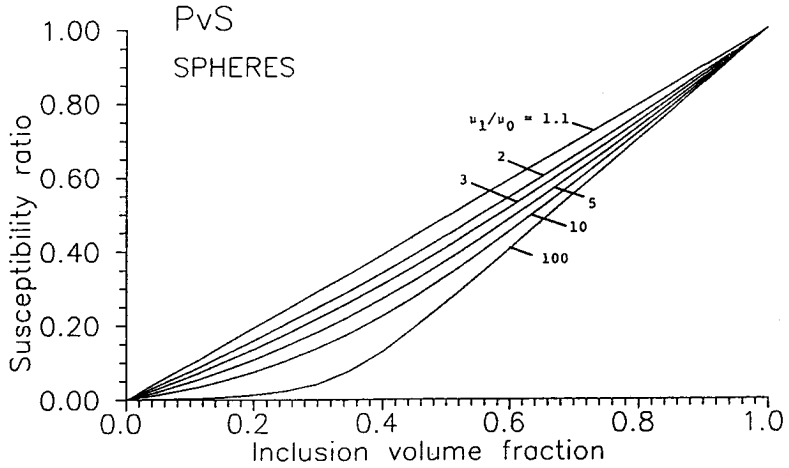


**Figure 4.4 Normalized effective permeability (susceptibility ratio $\mu_{\text{eff}} - \mu_0/\mu_1 - \mu_0$) of a two-phase mixture as a function of the volume fraction of the inclusion phase. The relative inclusion permeability is a parameter. Polder–van Santen formula with spherical inclusions.**

Using the expression for the magnetic polarizability (14), the effective permittivity can be given explicitly

$$\mu_{\text{eff}} = \mu_0 + 3\mu_0 \frac{\sum_{i=1}^{N} f_i \frac{\mu_i - \mu_0}{\mu_i + 2\mu_0}}{1 - \sum_{i=1}^{N} f_i \frac{\mu_i - \mu_0}{\mu_i + 2\mu_0}} \tag{51}$$

This is the multiphase magnetic Maxwell-Garnett mixing formula.

For mixtures with ellipsoidal inclusions the average magnetic polarization is a twofold sum: first, over the phases, and second, over the ellipsoid axial directions. The effective permeability for a mixture with $N$ ellipsoidal inclusion phases, each with given depolarization factors

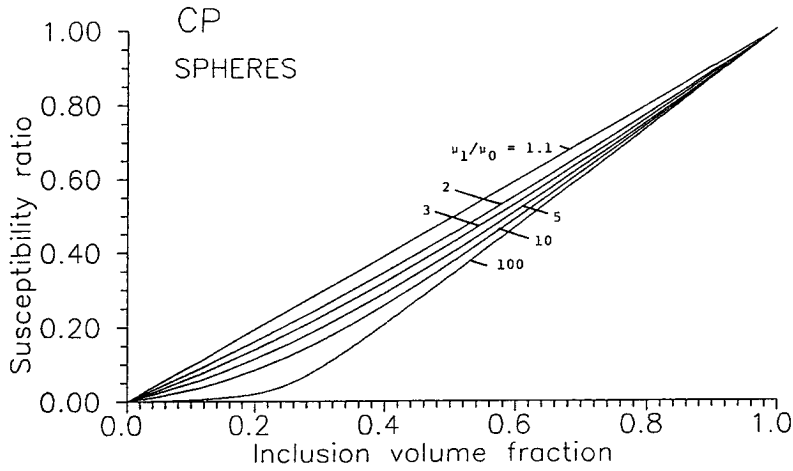


**Figure 4.5 Normalized effective permeability (susceptibility ratio $\mu_{\text{eff}} - \mu_0/\mu_1 - \mu_0$) of a two-phase mixture as a function of the volume fraction of the inclusion phase. The relative inclusion permeability is a parameter. CP formula with spherical inclusions.**

is a generalization of (35), and all inclusions randomly oriented is

$$\mu_{\text{eff}} = \mu_0 + \frac{\frac{1}{3}\mu_0 \sum_{j=1}^{N} f_j(\mu_j - \mu_0) \sum_{i=a,b,c} \frac{1}{\mu_0 + N_{ji}(\mu_j - \mu_0)}}{1 - \frac{1}{3}\sum_{j=1}^{N} f_j(\mu_j - \mu_0) \sum_{i=a,b,c} \frac{N_{ji}}{\mu_0 + (\mu_j - \mu_0)}} \tag{52}$$

where $N_{ji}$ is the depolarization factor of of the $j$th phase inclusion in the direction of the $i$th axis.

Even continuous distribution of the permeability in the inclusions can be treated: the sum over the $N$ phases will be replaced by an integral. However, care has to be taken in applying the multiphase mixing formula in the respect that it is valid only as long as the mixture is homogeneous when regarding scales of the order of wavelength: in

each volume unit of this size, there must be several inclusions of each particle phase.

Following the same averaging process for the magnetic polarization, also the Polder–van Santen formula and the CP formula can also be generalized to the multiphase mixture case.

Mixtures with inhomogeneous scatterers can also be analyzed. Considering (13), it is seen that the effective permeability of mixtures with spherical but not necessarily homogeneous inclusions is calculable provided that the magnetic polarizability of these inclusions is known. There are two ways to solve the polarizability of homogeneous as well as nonhomogeneous inclusions. First, solving the internal field inside the inclusion as it is immersed in a static field, allows the use of the definition of the dipole moment: volume integral of the product of the susceptibility and the field. The second approach is to solve the perturbative dipole field outside the inclusion due to its dipole moment, and from this field, the amplitude of the dipole moment can be enumerated. Both ways require solving the Laplace equation.

In [34], the Laplace equation for multilayer dielectric sphere is solved by transmission line analogy. The two types of allowed field solutions, a constant and a dipole type of field, are interpreted as radially forward- and backward propagating waves, as in a dynamical wave problem. The analysis is applicable, by duality, to the magnetic polarizability of a multilayer spherical inclusion. For example, the effective permeability of a mixture with magnetically coated spherical inclusions obeys the following formula (see [7,35]):

$$\frac{\mu_{\text{eff}} - \mu_0}{\mu_{\text{eff}} + 2\mu_0} = (f_1 + f_2) \frac{f_1(\mu_1 - \mu_0) + f_2 \dfrac{3\mu_1}{\mu_2 + 2\mu_1}(\mu_2 - \mu_0)}{f_1(\mu_1 + 2\mu_0) + f_2 \dfrac{3\mu_1}{\mu_2 + 2\mu_1}(\mu_2 + 2\mu_0)} \tag{53}$$

where $\mu_0$ is the background permeability, $\mu_1$ is the coating permeability, and $\mu_2$ is the permeability of the spherical core of the inclusions. $f_1$ and $f_2$ are the volume fractions of the coating material and the core material, respectively. Using this formula, each inclusion has to possess the same relative composition of materials $\mu_1$ and $\mu_2$, otherwise the numerator and the denominator of the mixing formula have to be modified to include sums of the different polarizabilities. Comparing (51) with $N = 2$ to (53), it is seen that even if the permeabilities and the volume fractions are the same, the effective permeability of the

mixture is different depending on the internal structure of the mixture; whether the phases are correlated (53) or uncorrelated (51). The nominators and denominators in (53) become lengthy for inclusions with increasing number of layers. However in [7], an explicit form is given to calculate the effective permittivity of a mixture with inclusions that may have any number of layers.

Dielectric mixtures with continuously inhomogeneous spherical scatterers have also been treated [7]. In the analysis of this reference, the permittivity function only depends on the radial distance from the core. Given the permittivity profile, the polarizability can be calculated from an ordinary differential equation. A variational method, which provides an approximate way for solving the polarizability [7], is also applicable to magnetic mixtures by duality. Multilayer and continuously inhomogeneous ellipsoids are analyzed in [36].

## 4.4 Limiting Formulas for Large Inclusion Permeability

In this section, the focus is on magnetic mixtures that possess large permeability contrasts, i.e. the permeability of one of the constituents is much larger than the permeabilities of the other constituents present in the mixture. Many ferromagnetic materials are within this category. However, care has to be exercised in analyzing the losses: for well-conducting inclusions, the penetration depth may be smaller than the size of the inclusions, and therefore the analysis in previous sections is not applicable. For small inclusions in this respect, the formulas are correct.

Consider a two-phase mixture with spherical high-permeability inclusions: $\mu_1 >> \mu_0$. From (15), the effective permeability is given by

$$\frac{\mu_{\text{eff}} - \mu_0}{\mu_{\text{eff}} + 2\mu_0} \simeq f \tag{54}$$

or

$$\mu_{\text{eff}} \simeq \mu_0 + \frac{3f\mu_0}{1-f} \tag{55}$$

which entails that for low volume filling ratios, $f << 1$, the effective permeability is approximately $\mu_{\text{eff}} \simeq \mu_0(1+3f)$. This is applicable for the real part $\mu'_{\text{eff}}$ of the effective permeability $\mu'_{\text{eff}} + i\mu''_{\text{eff}}$ but for enumerating the attenuation of the high-permeability contrast mixture with low loading in question, the approximation is too crude, because

magnetic loss information of the scatterers is lost in the approximation. If the magnetic loss of the mixture is included, the term $\frac{\mu_1 - \mu_0}{\mu_1 + 2\mu_0}$ has to be expanded into real and imaginary parts. If this is taken into account, the imaginary part of the effective permeability is

$$\mu''_{\text{eff}} \simeq f \left( \frac{3\mu_0}{\mu'_1} \right)^2 \mu''_1 \tag{56}$$

where $\mu'_1$ and $\mu''_1$ are the real and imaginary parts of the inclusion permeability $\mu_1$ and the assumption that $\mu''_1 << \mu'_1$ has been made.

Consider next mixtures with coated magnetic inclusions. Let the permeability of the coating be large compared to the background and the core. From (53), the effective permeability can be seen to be

$$\frac{\mu_{\text{eff}} - \mu_0}{\mu_{\text{eff}} + 2\mu_0} \simeq f_1 + f_2 \tag{57}$$

For multilayer scatterers coated with high-permeability layer, this equation can also be used; in this case, the sum on the right-hand side contains the volume fractions of all the layered constituents. This means that the outermost layer prevents the internal structure of the scatterer from having effect on the macroscopic permeability and the inclusion behaves as if it were completely of a high permeability. This phenomenon may be useful in designing materials that are to exhibit large permeability but are subject to weight restrictions. Again, care has to be exercised when considering losses. In [35], it is shown that melting hail (water-covered ice particles) may exhibit more attenuation than the same amount of precipitation in the form of rain although the amount of lossy material is larger in the latter case.

Figures 4.1, 4.4, and 4.5 reveal also interesting features from the point of view of high permeability contrast between the inclusion and the background. It can be seen that for high inclusion-background permeability ratio, the Polder–van Santen and the CP formulas exhibit a sudden increase of the effective permeability at volume fractions around 30%$\cdots$40%, a phenomenon that is absent in the Maxwell-Garnett case. This increase can be attributed to magnetic percolation analogous to the percolation phenomenon in certain dielectrics. The inability of the Maxwell-Garnett mixing formula to explain percolation would further support the view that its validity is restricted to sparse and low-contrast mixtures.

## 4.5 Discussion on the Losses and Nonlinearity

Within the techno-scientific remote sensing community, there has been discussion on the applicability of dielectric mixing formulas like the Maxwell-Garnett formula or the Polder–van Santen formula in the complex regime; i.e. if losses are included in the imaginary parts of the constituents of the component permittivities, does the imaginary part of the effective permittivity calculated from the mixing formula correctly represent the losses of the mixture? The same question can be raised concerning magnetic mixtures, and even with a more worried tenor, because the loss mechanisms in magnetic materials may be more complex.

The answer to this loss problem depends on the structure of the mixture, the size and form of the inclusions, and the loss mechanism itself. If the loss mechanism is the same in bulk material as in the case when this material is in the form of small particles, and the internal field calculation inherent in the derivation of the mixing formula is valid also for the lossy case, then also the imaginary part of the mixing formula correctly gives the mixture losses. This is the case in, for example, mixtures containing water, at microwave frequencies: the losses are of polar origin, and are due to the reaction of the polar water molecule to the incident field. As long as mixtures are concerned in which the inclusion dimensions are large compared with the molecular dimensions, the mixture loss calculation requires no special attention compared with the losses of homogeneous water.

It is worth noting that with the assumptions above, the mixing formula also takes into account the losses in the case that the inclusion phase is conducting. Consider a dielectric mixture where the background material is of permittivity $\epsilon_0$ and the inclusion phase has conductivity $\sigma_1$, and its complex permittivity is $\epsilon_1 = \epsilon_1' + i\sigma_1/\omega$. If the volume fraction of the inclusion phase $f$ is small, the effective conductivity $\sigma_{\text{eff}} = \omega \text{Im}\{\epsilon_{\text{eff}}\}$ of the mixture, calculated from the dielectric Rayleigh mixing formula, is

$$\sigma_{\text{eff}} = \frac{9\epsilon_0^2 f \sigma_1}{(\epsilon_1' + 2\epsilon_0)^2 + \dfrac{\sigma_1^2}{\omega^2}} \tag{58}$$

From this formula it is seen that the effective DC conductivity vanishes: $\sigma_{\text{eff}} \to 0$ as $\omega \to 0$. This is also intuitively clear: non-contacting conducting particles in a non-conducting matrix do not make the mixture

conducting. The same result is seen from the mixing formula derived by Wait [37] for the effective conductivity of a heterogeneous material (which is dual to the dielectric and magnetic problems).

Equation (58) shows that the loss factor and the imaginary part of the permittivity, that varies as $\omega^{-1}$ for the conductive inclusion material, is converted to the so called Maxwell-Wagner losses of the mixture (see for example [38]): losslessness in the low-frequency region, and normal conducting losses at high frequencies. The imaginary part of the effective permeability and the loss factor have their maximum value at the frequency

$$\frac{\omega}{2\pi} = \frac{1}{2\pi} \frac{\sigma_1}{\epsilon_1' + 2\epsilon_0} \tag{59}$$

This frequency is around 200 kHz for water of conductivity 1 mS/m.

Troubles in applying mixture formulas arise when the internal field of the inclusions, due to losses, is not calculable from the permittivity ratios as was done in the previous sections. This is the case for mixtures where the inclusions are large compared to the penetration depth of the field into the inclusions. Inside lossy inclusions, the field decays exponentially, and quick decay is not compatible with the algebraic calculation of the inside field. Consequently the mixing formula predicts too large internal field, and gives too large losses for the mixture, for example, in the cases of metal spheres with radius considerable larger than the penetration depth $\sqrt{2/\omega\mu\sigma}$.

In [4], three types of losses are distinguished that magnetic materials exhibit: eddy-current loss, after-effect loss, and hysteresis loss. Eddy-current loss is the conductivity loss considered above and it appears also in mixtures of magnetic materials. It is taken care of through treating the material as dielectric mixture with the conductivity included in the imaginary part of the inclusion permittivity, at the same time being aware of the penetration depth limitation discussed. Ferrites, however, are poor conductors compared to magnetic alloys, and the penetration depth is therefore large. Hence, for mixtures containing ferrites, this limitation is not severe. After-effect loss is due to the delay in the flux as the magnetic field changes suddenly. In [4], thermal fluctuation after-effect and diffusion after-effect are separated and discussed. However, as far as effective permeability of mixtures is concerned, the after-effect loss behaves similarly as the hysteresis

loss and, in addition, magnetic after effects are usually very small and would normally not be seen in experiments.

The hysteresis loss is related to the nonlinearity of the relation between the magnetic flux and the magnetic field. The magnetization of the material depends not only upon the magnetic field at that moment but also on the history of the magnetization. The principal characteristic of magnetic materials is the spontaneous alignment of the atomic moments parallel to each other, leading to high permeability. However, the parallel orientation is a local phenomenon, confined to magnetic regions called Weiss domains. The domains are separated from each other by Bloch walls. The incident magnetic field exerts a force on the magnetic dipole moments not parallel to the field direction and as the field intensity increases, the Bloch walls start to deform like elastic membranes, but the so called pinning points keep their places. As the field strength still increases, the pressure on the walls causes the pinning points to give way according to the local easy magnetization directions until finally saturation magnetization is reached. Changing the pinning points entails power consumption, which is the origin of hysteresis loss. However, in microwave applications, the magnetic field strength incident on materials is normally of the order of milliamperes per meter whereas the saturation fields and coercive fields in question as the nonlinearities of magnetic materials are discussed are of the order of hundreds of amperes per meter. Therefore, low-field approximations are justified, where the permeability can be considered of being a sum of a constant term and a small term linearly dependent on the magnetic field (see [4]). Still, it is possible to analyze the properties of composite magnetic materials for arbitrary fields, and calculate how the nonlinearity of the inclusion phase renders the effective permeability also nonlinear. In this case, the exciting field also depends on the history of the magnetization, i.e. the point on the hysteresis curve, and the hysteresis curve of the mixture is probably easiest to construct graphically from the hysteresis curve of the inclusion phase.

The mixture hysteresis losses also depend on the size of the inclusions with respect to the size of the Weiss domains. The Weiss domains can measure up to 1 mm. If the magnetic inclusions in a mixture are smaller than this, the hysteresis loss depends on the structure of the wall between the magnetic material and the background medium. It is unlikely in this case that any magnetic domain walls are generated in the magnetic reversal process unless the matrix is also strongly mag-

netic.

One important loss mechanism present in ferrites at microwave frequencies is that associated with the ferromagnetic resonance which is determined by the demagnetizing fields and anisotropy [39]. A little above the resonant frequency, the imaginary part of the permeability exhibits a maximum. Paté et al. have found [40] that the effect of ferromagnetic resonance shifts toward lower frequencies as the ferrite is mixed into a polymer matrix to form a composite material.

The temperature dependence of the magnetic properties of a mixture follows directly from the temperature dependence of the permeability of the inclusion material. From the mixing formula, the effective permeability in different temperatures $T$ can be calculated, which gives the $\mu_{\text{eff}}(T)$ dependence. However, without mixing rules it is easy to infer that above the Curie temperature of the inclusion material, also the mixture loses its permeability, provided that the temperature profile is constant in the mixture.

As has been emphasized in the analysis above, the quasistatic mixing formulae apply for mixtures where the inclusion sizes are considerably smaller than the wavelength. Also the characteristic size of the magnetic domain of a material in relation with the wavelength determines the frequency behavior of the permeability and magnetic losses, which might be different for bulk material and a mixture. If effective parameters at higher frequencies need be calculated than the quasistatic approach allows, one has to resort to scattering equations. The scattering effects become visible first in the imaginary part of the effective permeability, where in addition to magnetic losses contained in the imaginary parts of the constituent materials, an extra term comes due to the scattering losses.

Provided that the permeability values of a bulk material can be applied also for the inclusions, also the frequency dependence of the effective permeability can be found out by using the mixing formula and the frequency dependence of the permeabilities of the constituent materials. It is worth noting that a mixing formula may dramatically change the frequency behavior of the permeability of a mixture compared to that of the inclusion phase. Examples discussed above were the Maxwell-Wagner effect and the shift of ferromagnetic resonance in composite ferrite materials. One further striking example is the dielectric mixture of air and water: bulk water has a maximum of the imaginary part of the permittivity at the relaxation frequency, which

is around 9 GHz at the temperature of 0° C. The mixture of air and water, where water is in the form of small spheres (fog), also obeys the Debye frequency dependence typical of polar molecules, but its relaxation frequency is much larger, over 100 GHz. This is an opposite behavior compared to the shift toward lower frequencies of the ferromagnetic resonance in composite materials. The fourth example is the study of metal-semiconductor mixtures, where the Maxwell-Garnett formula has been found to explain the shifts of absorption and transmission peaks of gold-silica and silver-silica mixtures [41], as the correct frequency dependence of the permittivity model for metals has been applied.

## 4.6 Conclusion

This article has discussed magnetically heterogeneous media from the macroscopic point of view: how to describe the mixture in simple terms in scales of the magnetic field variation. The quantity in the focus of analysis is effective permeability, generally a complex number. Based on the decoupling of the magnetostatic and electrostatic problems and their duality, the dielectric mixing formulae are carried over to this permeability regime. Owing to the separability of the electrostatic and magnetostatic problems which can be seen from Maxwell's equations, the same mixing formula can be used to calculate both the effective permittivity and permeability. The real part of the effective permeability is fairly unproblematic and in principle, the losses can also be calculated from the mixing formula with complex permeability values. However, it is essential to be aware of the original physical mechanism behind the losses: whether the loss effect is the same in the bulk material as in the case when the material is in the form of small inclusions. Also, large losses need special attention: the penetration depth of the field must not be much smaller than the size of the scatterers. Another restriction that limits the size of the inclusions is the quasistatic analysis in the first place: compared to the wavelength, the inclusions need be small.

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