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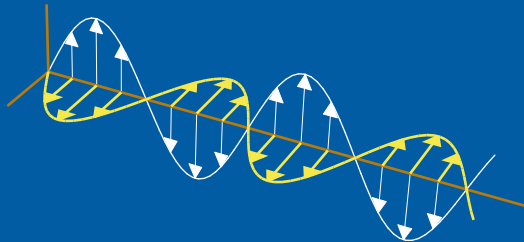
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Mixing formulas in time domain

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Abstract

This paper discusses the dispersive properties of dielectric materials both in the time and the frequency domains. Special emphasis is on the treatment of heterogeneous materials, in particular two-phase mixtures. A time domain Maxwell Garnett rule is derived which differs from the corresponding frequency domain formula in the respect that it is expressed in terms of convolutions and inverse operators of the susceptibility kernels of the materials. Much of the analysis deals with the question how the temporal dispersion of the dielectric response of various physical materials is affected by the mixing process. Debye, Lorentz, Drude, and modified Debye susceptibility models are treated in detail.

1 Introduction

The purpose of the present paper is twofold. One of the aims is to present a time domain approach to the effective-medium modeling of dielectrically heterogeneous materials. A second objective of the present analysis is to study how the mixing process affects the frequency behavior of temporally dispersive materials, which task will be mostly performed in the frequency domain. Because the time domain response of a medium is the primary mechanism that determines its frequency dispersion, these two aspects arise from the same physical basis.

One of the motivations for the time domain analysis to the homogenization problem is that to our knowledge it has not been performed previously. The classical mixing theories make use of static treatment of inclusions embedded in a host matrix and the effective permittivity is enumerated with the help of the polarizabilities of the inclusions (see, for example [3]). Alternatively, a long-wavelength limit of a time-harmonic dynamic problem is used to calculate the local fields and effective parameters of the mixture (see, for example [13]). A time domain approach differs from these analyses in the manner that the permittivity functions of the problem are convolution operators acting on the electric fields, and therefore a different algebraic formalism has to be applied to solve for the effective parameters of the mixture. The time domain response of the material is described by an instantaneous response and a susceptibility kernel responsible for the memory effects instead of frequency-dependent permittivity function as is customary in the frequency domain.

The two approaches are nevertheless connected through a Fourier transformation. Therefore, much of the properties of temporal dispersion of a dielectric mixture can be gleaned by using effective-medium theories in the frequency domain. It is astonishing how greatly the dielectric properties of a material in particulate form can differ from those of the bulk material. One is tempted to think that small particles are uninteresting and simple scatterers and absorbers and that they only repeat the spectral structure of the parent material, but this misconception is dangerous; on this issue the discussion in [2, Chap. 12] makes useful reading. For example, the absorption peaks in metals can be shifted in frequency when the material exists as inclusions, which is a well-known fact, and resonances can be generated by the mixing process that were not present in the response of the parent material. This paper

attempts to present a systematic analysis of the mixing effect on the dispersion, taking into account various fundamental dispersion models.

The present century has brought forward a large amount of effective-medium mixing models with always growing complexity in microstructure [15]. When taking the first step into the time domain of mixing rules it is reasonable to limit the analysis to the most basic model. In the following, the Maxwell Garnett model of a two-phase mixture is studied where the inclusions are embedded in the environment in the form of spheres, and both temporally dispersive materials are isotropic, homogeneous and non-magnetic.

The basic definitions and the constitutive relations are defined in Section 2. The Maxwell Garnett formula in the fixed frequency domain and in the time domain is presented in Section 3, and the dispersive effects in a mixture are analyzed in Section 4. Different dispersive models and mixtures illustrate the theory in Section 5. These results are presented in both the fixed frequency and in time domain domain. Conclusions and an appendix end the paper.

2 Definitions and constitutive relations

In the present paper we consider linear, isotropic, temporally dispersive dielectric materials. Therefore the medium displays neither magnetic susceptibility nor magnetoelectric coupling. The constitutive relations have a general form for such kind of medium, see [10]:

$$\begin{cases} \frac{1}{\varepsilon_0} \mathbf{D}(\mathbf{r}, t) = \varepsilon_\infty(\mathbf{r}) \mathbf{E}(\mathbf{r}, t) + (\chi * \mathbf{E})(\mathbf{r}, t) \\ \mathbf{B}(\mathbf{r}, t) = \mu_0 \mathbf{H}(\mathbf{r}, t) \end{cases} \quad (2.1)$$

where the vacuum permittivity is ε_0 and the vacuum permeability μ_0 .

The electric and magnetic fields $\mathbf{E}, \mathbf{D}, \mathbf{H}, \mathbf{B}$ appearing in these constitutive relations have an explicit space and time dependence. Since we are analyzing the dielectric material at a given fixed position, the spatial dependence is suppressed for convenience. Of these two constitutive equations, the magnetic relation is trivial. The dielectric relation is separated into two parts: the optical response and a dispersive part. The function $\varepsilon_\infty(\mathbf{r}) \geq 1$ describes the instantaneous (optical) response of the material. The dispersive part is described by the susceptibility kernel $\chi(\mathbf{r}, t)$ as a function of time, and the operation denoted by $*$ stands for the temporal convolution:

$$(\chi * \mathbf{E})(t) = \int_{-\infty}^t \chi(t - t') \mathbf{E}(t') dt'$$

The susceptibility kernel is assumed to have piecewise continuous derivatives for $t > 0$. Due to causality, the susceptibility kernel χ vanishes for $t < 0$, but it can have a discontinuity in the origin, *i.e.*, it can happen that $\chi(t = 0^+) \neq 0$. Although this option of discontinuity across the origin is sometimes doubted in the literature (see, for example [8, p. 310], [18]), it does not violate causality.

In the following, we shall treat the permittivity functions using both their time domain and frequency domain descriptions. On the level of notation, we try to distinguish the operator form of the permittivity by denoting it with ϵ whereas any permittivity quantity not depending on time is written as ε , be it constant or depending on frequency. Note that both the permittivity operators and constants are dimensionless, relative quantities, and the susceptibility kernel has the dimension of frequency.

The frequency domain counterpart of the dielectric constitutive relation in (2.1) is

$$\frac{1}{\varepsilon_0} \mathbf{D}(\omega) = \varepsilon(\omega) \mathbf{E}(\omega)$$

where the Fourier transformation of the quantities are defined by the following convention

$$\left\{ \begin{array}{l} \mathbf{D}(\omega) = \int_{-\infty}^{\infty} \mathbf{D}(t) e^{-j\omega t} dt \\ \mathbf{E}(\omega) = \int_{-\infty}^{\infty} \mathbf{E}(t) e^{-j\omega t} dt \\ \varepsilon(\omega) - \varepsilon_{\infty} = \int_{-\infty}^{\infty} \chi(t) e^{-j\omega t} dt \end{array} \right. \quad (2.2)$$

In the frequency domain, the Kramers–Kronig relations are fulfilled for causal materials that have an absolutely integrable susceptibility kernel [8, pp. 309-312].

3 Maxwell Garnett formula

The objective of mixing rules is to connect the macroscopic effective properties of a heterogeneous medium to its structural parameters: volume fractions of the constituents and their dielectric properties. Consider a mixture of two isotropic homogeneous dielectric materials of type (2.1). Let one of those form the background matrix and let the other be a “guest phase” in the form of spheres, and occupying a volume fraction f of the total space, as illustrated in Figure 1.

3.1 Frequency domain

In the frequency domain characterization of dielectric materials, we describe the material with the permittivity function $\varepsilon(\omega)$. Let us denote the dielectric permittivity of the background medium by ε_b and that of the inclusions by ε_i , and both quantities are understood to be functions of frequency in general. According to the Maxwell Garnett formula [5], the effective permittivity of the mixture with spherical inclusions is

$$\varepsilon_{\text{eff}} = \varepsilon_b + 3f\varepsilon_b \frac{\varepsilon_i - \varepsilon_b}{\varepsilon_i + 2\varepsilon_b - f(\varepsilon_i - \varepsilon_b)} \quad (3.1)$$

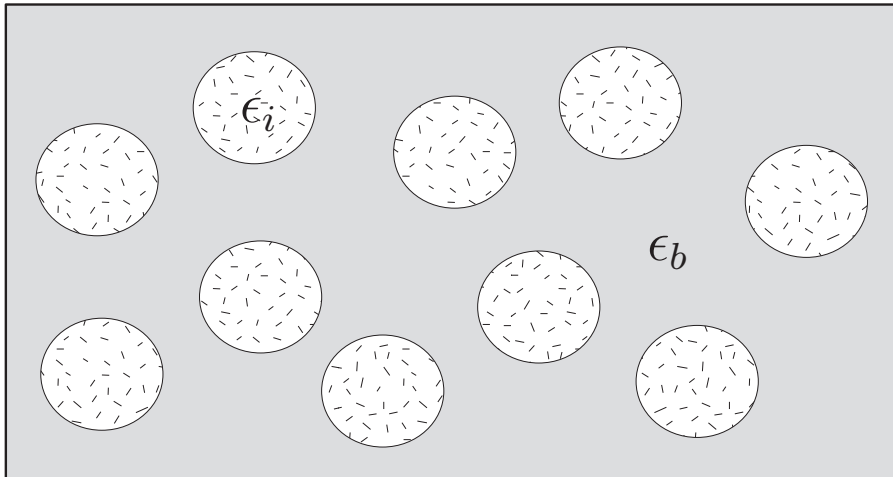


Figure 1: The geometry of the mixing problem: spherical isotropic dielectric inclusions in isotropic dielectric environment.

For sparse mixtures, *i.e.*, the inclusion phase is very dilute ($f \ll 1$), the Maxwell Garnett (MG) rule reads

$$\varepsilon_{\text{eff}} = \varepsilon_b + 3f\varepsilon_b \frac{\varepsilon_i - \varepsilon_b}{\varepsilon_i + 2\varepsilon_b} \quad (3.2)$$

Despite its simplicity, the Maxwell Garnett rule is much used and also a successful model. Let us try to formulate this formula with quantities having explicit time dependence.

3.2 Time domain

If the mixing problem is treated in the time domain, the fields and flux densities have explicit time dependence. The time-dependent polarizability characteristics of the material inclusions need to be calculated. Consider the permittivity operators of the materials, denoted by ϵ and defined in the following way:

$$[\epsilon \mathbf{E}](t) = [(\varepsilon_\infty + \chi^*) \mathbf{E}](t) = [(\varepsilon_\infty \delta + \chi) * \mathbf{E}](t) \quad (3.3)$$

where the variable \mathbf{r} has been suppressed and $\delta(t)$ is the Dirac delta-function. Note that the convolution asterisk is contained within the permittivity operator ϵ .

Now the problem of a spherical inclusion (permittivity operator ϵ_i) in a homogeneous background medium (permittivity operator ϵ_b) can be formulated. To solve the dipole moment induced in the inclusion, the internal field needs to be known first. Then the dipole moment operator can easily be calculated as an integral of the polarization density over the inclusion volume.

The homogenization of a mixture is always an approximation because the number of parameters of the structure is being decreased considerably. High-frequency scattering effects from a heterogeneous medium cannot be explained by modeling the medium effectively homogeneous and therefore the macroscopic permittivity has

only a meaning if the sources and the fields vary temporally in a sufficiently slow manner. This is a limitation that has to be encountered both in the frequency and the time domains.

The quasistatic limitation in the frequency domain is often expressed by saying that the size of the scatterer should be small in comparison with the wavelength of the field. However, in the time domain formulation there are no wavelengths. Instead, the corresponding limitation relates the general time variation of the source with the propagation velocity v and the size d of the inclusions. When the “temporal size” $\Delta t = d/v$ of the inclusion is small compared with the inverse of the time derivative of the source, we can consider the field inside an inclusion homogeneous even if the field varies temporally. Time only appears as a parameter in the field expressions.

Although the retardation effects are excluded within the homogenization analysis, memory effects are allowed in the dielectric materials. These are contained in the susceptibility kernels. The high-frequency components of the material response are integrated into the optical response (delta-function) part of the representation (3.3).

To calculate the internal field, one can follow the steps of the classical analysis of the sphere in a uniform and static field (*cf.*, for example [8, Sect. 4.4]). The relation between the internal field $\mathbf{E}_i(t)$ and the external field $\mathbf{E}_e(t)$ comes as

$$[(\epsilon_i + 2\epsilon_b)\mathbf{E}_i](t) = 3[\epsilon_b\mathbf{E}_e](t)$$

This equation looks formally similar to the purely static result but attention must be paid to the fact that the permittivities are here convolution operators. Hence, to solve for the internal field, a deconvolution has to be performed:

$$\mathbf{E}_i(t) = [(\epsilon_i + 2\epsilon_b)^{-1}3\epsilon_b\mathbf{E}_e](t)$$

The deconvolution for the operators of the type (3.3) can be done provided that the optical response of the operator is nonzero. The detailed calculation of the inverse is discussed in Appendix A.

The polarizability operator for a single inclusion, α , being defined through the dipole moment $\mathbf{p}(t) = [\alpha\mathbf{E}_e](t)$ can now be written as

$$\alpha = \epsilon_0 V (\epsilon_i - \epsilon_b) (\epsilon_i + 2\epsilon_b)^{-1} 3\epsilon_b \quad (3.4)$$

where V is the volume of the sphere.

For a mixture, the exciting field that has to be used to calculate the dipole moment is the so-called Lorentzian field [19]. In time domain description this field looks like:

$$\mathbf{E}_L(t) = \mathbf{E}_e(t) + \frac{1}{3\epsilon_0} [\epsilon_b^{-1}\mathbf{P}](t)$$

where $\mathbf{P} = n\mathbf{p}$ is the average polarization in the mixture (n is the number density of inclusions). Using now the Lorentzian field on which the polarizability operator

acts to calculate the dipole moment ($\mathbf{p} = \alpha \mathbf{E}_L$), we can solve the Lorentzian field in terms of the average field:

$$\mathbf{E}_L(t) = \left[\left(3\epsilon_b - \frac{n\alpha}{\epsilon_0} \right)^{-1} 3\epsilon_b \mathbf{E}_e \right] (t)$$

which gives us the effective permittivity operator, being defined by $\epsilon_0 [\epsilon_{\text{eff}} \mathbf{E}_e] (t) = \epsilon_0 \epsilon_b \mathbf{E}_e(t) + \mathbf{P}(t)$:

$$\epsilon_{\text{eff}} = \epsilon_b + \frac{n\alpha}{\epsilon_0} \left(3\epsilon_b - \frac{n\alpha}{\epsilon_0} \right)^{-1} 3\epsilon_b \quad (3.5)$$

This could be termed as the time domain Lorenz–Lorentz formula¹ because it gives the effective permittivity as a function of the polarizability operator [15].

For a sparse mixture, the norm of the operator $n\alpha/\epsilon_0$ is much smaller than that of $3\epsilon_b$, which means that the effective permittivity is

$$\epsilon_{\text{eff}} = \epsilon_b + \frac{n\alpha}{\epsilon_0} = \epsilon_b + f(\epsilon_i - \epsilon_b)(\epsilon_i + 2\epsilon_b)^{-1} 3\epsilon_b$$

where $f = nV$ is the volume fraction of the inclusion phase in the mixture.

The full Lorenz–Lorentz formula (3.5) can also be manipulated further. Noting that the convolution operator commutes:² $(a + A*)(b + B*) = (b + B*)(a + A*)$, and for piecewise continuous kernels, the inverse of the operator product is

$$[(a + A*)(b + B*)]^{-1} = (a + A*)^{-1}(b + B*)^{-1}$$

we can write the time domain Lorenz–Lorentz formula in a Maxwell Garnett form

$$\epsilon_{\text{eff}} = \epsilon_b + f(\epsilon_i - \epsilon_b)[\epsilon_i + 2\epsilon_b - f(\epsilon_i - \epsilon_b)]^{-1} 3\epsilon_b \quad (3.6)$$

Note that in the derivation of the polarizability, (3.4), we have assumed that the optical response of the operator $\epsilon_i + 2\epsilon_b$ has to be nonzero. This is a reasonable assumption because its violation would correspond to the unphysical condition

$$\lim_{\omega \rightarrow \infty} \frac{\epsilon_i(\omega)}{\epsilon_b(\omega)} = -2$$

Furthermore, the evaluation of the expression (3.6) requires that the optical response of $\epsilon_i + 2\epsilon_b - f(\epsilon_i - \epsilon_b)$ must not vanish.

Let us next move back into the frequency domain to analyze the dispersive characteristics of a mixture in more detail.

¹“Time domain Clausius–Mossotti formula” is equally suitable.

²Here a and b are nonzero constants.

4 Modeling dispersive effects

The starting point in this section are the permittivity functions ε_i and ε_b modeled as a general quotient of two polynomials.

$$\varepsilon_i(\omega) = \frac{\sum \alpha_m^i \omega^m}{\sum \beta_n^i \omega^n}$$

and

$$\varepsilon_b(\omega) = \frac{\sum \alpha_m^b \omega^m}{\sum \beta_n^b \omega^n}$$

The limits of the summation in the numerator and the denominator are suppressed for convenience. However, it is assumed that all summations are finite and that only non-negative m - and n -values contribute. In most practical situations the degree of the polynomial in the nominator is less than in the denominator. The quotients are normalized by requiring $\beta_0^i = \beta_0^b = 1$.³

Both the sparse and the dense formulae, (3.1) and (3.2) can be written in the following form:

$$\varepsilon_{\text{eff}} = \varepsilon_b + 3f\varepsilon_b \frac{\varepsilon_i - \varepsilon_b}{\gamma_i \varepsilon_i + \gamma_b \varepsilon_b}$$

where the values for a full Maxwell Garnett formula are $\gamma_i = 1 - f$ and $\gamma_b = 2 + f$. For a sparse mixture the corresponding values are $\gamma_i = 1$ and $\gamma_b = 2$.

The general frequency behavior for ε_{eff} can be written as a quotient between two polynomials.

$$\varepsilon_{\text{eff}}(\omega) = \frac{\sum \alpha_m^e \omega^m}{\sum \beta_n^e \omega^n}$$

where

$$\left\{ \begin{array}{l} \alpha_m^e = \frac{\sum_{k,l} \alpha_k^b \{ \alpha_l^i \beta_{m-k-l}^b (\gamma_i + 3f) + \alpha_l^b \beta_{m-k-l}^i (\gamma_b - 3f) \}}{\alpha_0^i \gamma_i + \alpha_0^b \gamma_b}, \quad m = 0, 1, 2, 3, \dots \\ \beta_0^e = 1 \\ \beta_n^e = \frac{\sum_{k,l} \beta_k^b \{ \alpha_l^i \beta_{n-k-l}^b \gamma_i + \alpha_l^b \beta_{n-k-l}^i \gamma_b \}}{\alpha_0^i \gamma_i + \alpha_0^b \gamma_b}, \quad n = 1, 2, 3, \dots \end{array} \right. \quad (4.1)$$

If the background medium is non-dispersive, *i.e.*, ε_b is constant with respect to the frequency, the general expression (4.1) simplify. In this case it is easy to identify the coefficients α_m^e and β_n^e in the following expansions. The explicit expressions of

³If the denominator vanishes as $\omega \rightarrow 0$, *i.e.*, either β_0^i or $\beta_0^b = 0$, normalize such that the first non-vanishing coefficient β_n^i or $\beta_n^b = 1$. An example where this situation occur is in the Drude model which we analyze below.

the coefficients are:

$$\begin{cases} \alpha_m^e = \frac{\varepsilon_b(\gamma_i + 3f)\alpha_m^i + \varepsilon_b^2(\gamma_b - 3f)\beta_m^i}{\gamma_i\alpha_0^i + \varepsilon_b\gamma_b}, & m = 0, 1, 2, 3, \dots \\ \beta_0^e = 1 \\ \beta_n^e = \frac{\gamma_i\alpha_n^i + \varepsilon_b\gamma_b\beta_n^i}{\gamma_i\alpha_0^i + \varepsilon_b\gamma_b}, & n = 1, 2, 3, \dots \end{cases}$$

For a sparse mixture ($\gamma_i = 1$, $\gamma_b = 2$) we have

$$\begin{cases} \alpha_m^e = \frac{\varepsilon_b(1 + 3f)\alpha_m^i + \varepsilon_b^2(2 - 3f)\beta_m^i}{\alpha_0^i + 2\varepsilon_b}, & m = 0, 1, 2, 3, \dots \\ \beta_0^e = 1 \\ \beta_n^e = \frac{\alpha_n^i + 2\varepsilon_b\beta_n^i}{\alpha_0^i + 2\varepsilon_b}, & n = 1, 2, 3, \dots \end{cases}$$

and for the full Maxwell Garnett formula ($\gamma_i = 1 - f$, $\gamma_b = 2 + f$) we have

$$\begin{cases} \alpha_m^e = \frac{\varepsilon_b(1 + 2f)\alpha_m^i + 2\varepsilon_b^2(1 - f)\beta_m^i}{(1 - f)\alpha_0^i + \varepsilon_b(2 + f)}, & m = 0, 1, 2, 3, \dots \\ \beta_0^e = 1 \\ \beta_n^e = \frac{(1 - f)\alpha_n^i + \varepsilon_b(2 + f)\beta_n^i}{(1 - f)\alpha_0^i + \varepsilon_b(2 + f)}, & n = 1, 2, 3, \dots \end{cases} \quad (4.2)$$

The time domain expressions are very complex, unless the order of the polynomial is low. The explicit time domain results are best illustrated by the examples given in Section 5.

5 Examples of dispersive models and mixtures

Let us use the preceding results to study how the mixing process affects the characteristics of certain specific dispersion models: Debye, Lorentz, Drude, and modified Debye models.⁴ Let us first assume that the background material is non-dispersive, and that only the inclusions show dispersion. The general case of a mixture of two dispersive phases is then illustrated. The examples are first presented in the fixed frequency domain and then in the time domain.

5.1 Frequency domain

5.1.1 Debye model

The Debye model is suitable to describe the dielectric properties of fluids that have permanent electric dipole moments. The frequency-dependent permittivity function

⁴For more detailed discussion on the physical background of these models, see for example [2, Chap. 9], [11, Sect. 2.1], [17, Chap. 3].

for the Debye model reads

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + j\omega\tau} \quad (5.1)$$

where ε_s and ε_∞ are the low-frequency and optical permittivities of the material and τ is the relaxation time, respectively.⁵ The material is lossy (the time-harmonic convention $\exp(j\omega t)$ is assumed), and the dissipativity of the material is seen from the fact that the imaginary part of $\varepsilon(\omega)$ is negative for all frequencies. The imaginary part has its minimum⁶ at the relaxation frequency $f_r = 1/(2\pi\tau)$.

Suppose now that the background permittivity is a dispersionless constant ε_b and the frequency dependence of the inclusions follows the Debye law (5.1). Then the Maxwell Garnett prediction (3.1) for the mixture permittivity can be calculated, and it is

$$\varepsilon_{\text{eff}}(\omega) = \varepsilon_{\infty,\text{eff}} + \frac{\varepsilon_{s,\text{eff}} - \varepsilon_{\infty,\text{eff}}}{1 + j\omega\tau_{\text{eff}}}$$

In other words, the mixture also a Debye material. The parameters of the mixture are, (4.2)

$$\begin{cases} \varepsilon_{\infty,\text{eff}} = \varepsilon_b + 3f\varepsilon_b \frac{\varepsilon_\infty - \varepsilon_b}{\varepsilon_\infty + 2\varepsilon_b - f(\varepsilon_\infty - \varepsilon_b)} \\ \varepsilon_{s,\text{eff}} = \varepsilon_b + 3f\varepsilon_b \frac{\varepsilon_s - \varepsilon_b}{\varepsilon_s + 2\varepsilon_b - f(\varepsilon_s - \varepsilon_b)} \\ \tau_{\text{eff}} = \tau \frac{(1-f)\varepsilon_\infty + (2+f)\varepsilon_b}{(1-f)\varepsilon_s + (2+f)\varepsilon_b} \end{cases} \quad (5.2)$$

As an example of such a mixture let us treat water and air. For the temperature dependence of the dielectric properties of liquid water we take the following model [4]:

$$\varepsilon_s = 190.0 - 0.375 T, \quad \varepsilon_\infty = 4.90, \quad \tau = \frac{1.99}{T} e^{2140/T} \cdot 10^{-12} \text{ s}$$

where T is the temperature in Kelvin-degrees. See also [1, 9] for more information about the temperature dependence of the relaxation parameters of liquid water.

Using the result (5.2) for water drops in air ($\varepsilon_b = 1$), we can see that the relaxation frequency of the mixture depends strongly on the volume fraction of the water phase, and is shown in Figure 2. This figure shows, for example at +20° degrees Centigrade, that the relaxation frequency which for bulk water is around 17 GHz, is about 10 times higher for a dilute water-air mixture. Note also the very strong temperature dependence of the relaxation frequency τ_{eff} which comes from the sensitivity of the properties of bulk water on temperature.

In the real world, an example of this type of a mixture is a cloud or fog. Rain would also suggest itself, but a problem with raindrops is that they are not small in wavelengths at this interesting regime of millimeter wave frequencies, and therefore one cannot disregard the scattering effects.

⁵For real materials, this model is naturally valid only across a certain frequency band.

⁶Since $\text{Im}\{\varepsilon\}$ is negative, its absolute value has a maximum.

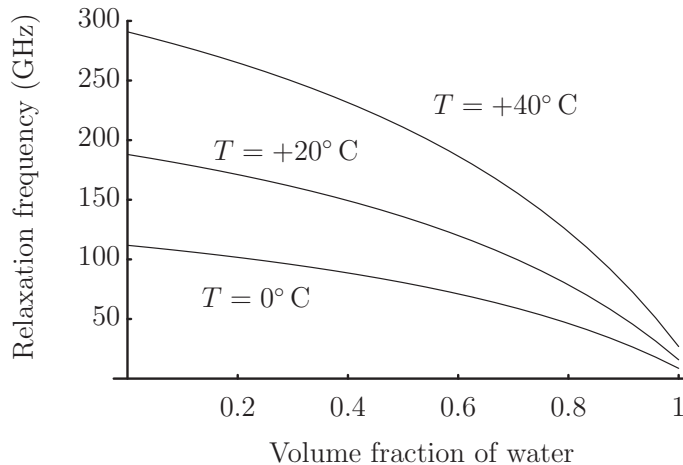


Figure 2: The relaxation frequency of water-air mixture as a function of the volume fraction of the water phase. The curves correspond to three different temperatures: 0° C, 20° C, and 40° C.

However, fog and clouds consist of water droplets having a size around micrometers, which assures that certainly below 500 GHz the scattering effects are negligible compared to absorption phenomena. Broadband microwave and millimeter wave propagation experiments have been performed to study the attenuation of fog and clouds [6, 16], and indeed the maximum attenuation has been observed to be around 200 GHz although the measurements are often impeded by additional attenuation factors in the atmosphere, like the absorption peaks of water vapor and other molecules present in the troposphere.

The time domain analysis of the same Debye mixture is given later in Section 5.2.1.

5.1.2 Lorentz model

The Lorentz model is a widely used model in solid state physics, and it predicts the frequency dependence of the permittivity function as

$$\varepsilon(\omega) = \varepsilon_{\infty} + \frac{\omega_p^2}{\omega_0^2 - \omega^2 + j\omega\nu} \quad (5.3)$$

where again ε_{∞} is the high-frequency permittivity of the material. The other parameters are the plasma frequency ω_p , the resonance frequency ω_0 , and the damping amplitude ν , also with dimensions of frequency.

The use of the Maxwell Garnett rule (4.2) shows that a mixture with Lorentz material (5.3) in dispersionless background medium, ε_b , is also a Lorentz-material,

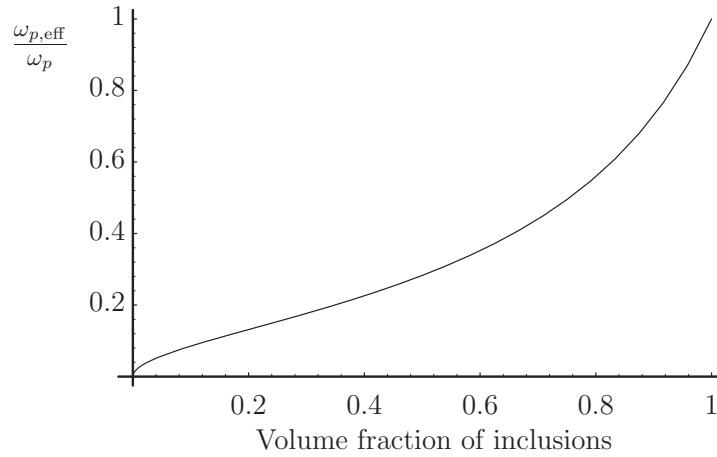


Figure 3: The plasma frequency of a mixture (relative to the plasma frequency of inclusions) with Lorentz spheres in air as a function of the volume fraction of the water phase. The high-frequency response is assumed to be $\varepsilon_\infty = 10$.

but the model parameters are transformed to

$$\begin{cases} \varepsilon_{\infty,\text{eff}} = \varepsilon_b + 3f\varepsilon_b \frac{\varepsilon_\infty - \varepsilon_b}{\varepsilon_\infty + 2\varepsilon_b - f(\varepsilon_\infty - \varepsilon_b)} \\ \omega_{p,\text{eff}} = \sqrt{f} \frac{3\varepsilon_b}{(1-f)\varepsilon_\infty + (2+f)\varepsilon_b} \omega_p \\ \omega_{0,\text{eff}}^2 = \omega_0^2 + \frac{1-f}{(1-f)\varepsilon_\infty + (2+f)\varepsilon_b} \omega_p^2 \\ \nu_{\text{eff}} = \nu \end{cases} \quad (5.4)$$

These results show that a mixture has a higher resonance frequency than the inclusion phase, and the shift of $\omega_{0,\text{eff}}$ from ω_0 is largest for dilute mixtures. Another observation is that the plasma frequency of a mixture increases for higher concentrations. This is natural because ω_p in fact is a measure of the permittivity magnitude as can be seen from (5.3). The damping factor ν is not affected by mixing.

Figure 3 shows the effect of mixing ratio on the plasma frequency of the mixture and Figure 4 displays the effect for the resonance frequency.

5.1.3 Drude model

A special case of the Lorentz model is the Drude model, used to describe the optical properties of metals. It comes from (5.3) by letting the electrons to be free, *i.e.*, assuming the resonance frequency is zero, $\omega_0 = 0$:

$$\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 - j\omega\nu} \quad (5.5)$$

The typical conductivity behavior for low frequencies

$$\varepsilon(\omega) \rightarrow -\frac{j\sigma}{\omega\varepsilon_0}$$

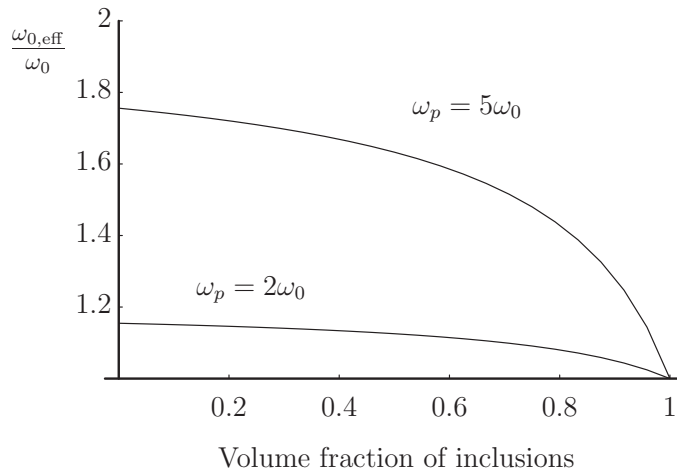


Figure 4: The resonance frequency of a mixture (relative to the resonance frequency of inclusions) with Lorentz spheres in air as a function of the volume fraction of the water phase. The high-frequency response is assumed to be $\varepsilon_\infty = 10$ and shown are two plasma-to-resonance frequency ratios of the inclusion phase.

is apparent in this model, where the conductivity is $\sigma = \omega_p^2 \varepsilon_0 / \nu$.

The result (4.2) shows now that the Maxwell Garnett mixture of Drude spheres in a dispersionless environment is a Lorentz model, where $\varepsilon_{\infty,\text{eff}}$, $\omega_{p,\text{eff}}$, and ν are identical to the Lorentz mixture, (5.4), but $\omega_{0,\text{eff}}$ is different. This means that the mixture does not follow a metal-type Drude model but an insulator-type resonator model (5.3). This is understandable because separate metal particles do not form a conducting lattice and therefore there is no low-frequency divergence of the permittivity. The mixture permittivity has a resonance frequency:

$$\omega_{0,\text{eff}} = \sqrt{\frac{1-f}{(1-f)\varepsilon_\infty + (2+f)\varepsilon_b}} \omega_p \quad (5.6)$$

For dilute mixtures and vanishing high-frequency response of the metal ($\varepsilon_\infty = 1$) this condition reads

$$\omega_{0,\text{eff}} = \frac{\omega_p}{\sqrt{1+2\varepsilon_b}} \quad (5.7)$$

Metal colloids have been studied extensively and indeed the condition (5.7) has been observed to hold for many metals like sodium, aluminum, and gold [2, 11]. The experiments show that a strong peak occurs for the absorption cross section of small spherical metal particles at this frequency $\omega_{0,\text{eff}}$.

The condition that determines the point of strong absorption for the small spherical metal inclusions carries also other names in the literature: terms like surface mode, surface plasmon, or Fröhlich frequency are associated with it. The studies dealing with plasmons and Fröhlich modes do not consider mixing rules but rather the scattering and absorption coefficients of inclusions. Looking at the polarizability

of a sphere,

$$\alpha = \varepsilon_0 V(\varepsilon_i - \varepsilon_b) \frac{3\varepsilon_b}{\varepsilon_i + 2\varepsilon_b}$$

it is then obvious that for a frequency at which $\varepsilon_i = -2\varepsilon_b$ something catastrophic happens, and the amplitude of the resulting absorption peak is determined by the imaginary part of the inclusion permittivity, assuming the background to be real. This condition corresponds to the sparse-mixture limit of the more general resonance condition (5.6).

5.1.4 Modified Debye model

The following model

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{\omega_p^2}{(\omega_0 + j\omega)^2} \quad (5.8)$$

is called the modified Debye model, and where ω_p and ω_0 are the plasma and resonance frequencies, respectively. The reason for the name is that the time domain susceptibility of the modified Debye model has the same exponential damping character as the Debye model but, unlike in the Debye model case, the initial value of the kernel vanishes (in the Debye model $\chi(0+) \neq 0$). See Section 5.2 for more details.

A mixture with modified Debye material embedded in non-dispersive host, ε_b , obeys the following frequency-dependent permittivity function:

$$\varepsilon_{\text{eff}}(\omega) = \varepsilon_{\infty, \text{eff}} + \frac{\omega_{p, \text{eff}}^2}{\omega_{0, \text{eff}}^2 - \omega^2 + j\omega\nu_{\text{eff}}}$$

where

$$\left\{ \begin{array}{l} \varepsilon_{\infty, \text{eff}} = \varepsilon_b + 3f\varepsilon_b \frac{\varepsilon_\infty - \varepsilon_b}{\varepsilon_\infty + 2\varepsilon_b - f(\varepsilon_\infty - \varepsilon_b)} \\ \omega_{p, \text{eff}} = \sqrt{f} \frac{3\varepsilon_b}{(1-f)\varepsilon_\infty + (2+f)\varepsilon_b} \omega_p \\ \omega_{0, \text{eff}}^2 = \omega_0^2 + \frac{1-f}{(1-f)\varepsilon_\infty + (2+f)\varepsilon_b} \omega_p^2 \\ \nu_{\text{eff}} = 2\omega_0 \end{array} \right.$$

It is seen that the mixture is no longer a modified Debye material, but a more general Lorentz material. The effective parameters $\varepsilon_{\infty, \text{eff}}$, $\omega_{p, \text{eff}}$, $\omega_{0, \text{eff}}$ obey the same rules as a mixture with Lorentz inclusions. This is obvious, if the denominator in (5.8) is expanded and compared with the form of the Lorentz model, (5.3). A difference in the mixing process is the appearance of a damping factor, which in the present case is twice the resonance frequency.

5.1.5 A mixture of two dispersive materials

If the background material is dispersive as well, the dispersion of the mixture is in general more complicated than that of the inclusion phases. The general frequency

behavior was derived in Section 4, see (4.1). To illustrate the difficulties, let us rewrite the permittivity functions as follows:

$$\begin{cases} \varepsilon_i(\omega) = \varepsilon_{\infty,i} + \Omega_i(\omega) \\ \varepsilon_b(\omega) = \varepsilon_{\infty,b} + \Omega_b(\omega) \end{cases}$$

where the frequency-dependent parts of the inclusions, $\Omega_i(\omega)$, and of the environment, $\Omega_b(\omega)$, vanish for high frequencies. It is here assumed that both the inclusions and the background have a high frequency limit $\varepsilon_{\infty,i}$ and $\varepsilon_{\infty,b}$, respectively. Then the Maxwell Garnett prediction, (3.1), for the mixture permittivity can be written as

$$\varepsilon_{\text{eff}}(\omega) = \varepsilon_{\infty,\text{eff}} + \Omega_{\text{eff}}(\omega)$$

where

$$\Omega_{\text{eff}} = \Omega_b + 3f \frac{3\varepsilon_{\infty,b}^2(\Omega_i - \Omega_b) + (1-f)(\varepsilon_{\infty,i} - \varepsilon_{\infty,b})^2\Omega_b + A(\Omega_i - \Omega_b)\Omega_b}{A[A + (1-f)\Omega_i + (2+f)\Omega_b]}$$

where $A = (1-f)\varepsilon_{\infty,i} + (2+f)\varepsilon_{\infty,b}$, and

$$\varepsilon_{\infty,\text{eff}} = \varepsilon_{\infty,b} + 3f\varepsilon_{\infty,b} \frac{\varepsilon_{\infty,i} - \varepsilon_{\infty,b}}{(1-f)\varepsilon_{\infty,i} + (2+f)\varepsilon_{\infty,b}} \quad (5.9)$$

This expressions show that even for the simplest case of mixing dispersive materials — Debye inclusions in Debye background — the effective permittivity is not of Debye form, not even a Lorentz form, but contains higher powers of ω both in the numerator and the denominator. One has to resort to the formulas in Section 4 to evaluate the required coefficients.

As an example, we analyze in the following the mixture of water and alcohol (ethanol). Both liquids have the Debye-type of permittivity behavior. We take the values at 20° C, whence the parameters are for water

$$\varepsilon_s = 80.1, \quad \varepsilon_{\infty} = 4.9, \quad \tau = 1.01 \cdot 10^{-11} \text{ s}$$

and those of ethanol

$$\varepsilon_s = 25.1, \quad \varepsilon_{\infty} = 4.4, \quad \tau = 1.2 \cdot 10^{-10} \text{ s}$$

Note the 12 times lower relaxation frequency for alcohol compared to liquid water. An informative illustration of the complex permittivity of materials is the so-called Cole–Cole plot where the permittivity curve is shown in the complex ε plane. For a Debye material, the Cole–Cole diagram is a semi-circle where relaxation frequency corresponds to the mid-point on the top of the half-circle.⁷

We show the Cole–Cole diagrams of two mixtures: alcohol–in–water and water–in–alcohol, and for both mixtures the inclusion volume fraction is 20%. Figure 5 shows the first case (water as background, spherical ethanol inclusions) and Figure 6 shows the second case (ethanol as background, inclusions as water drops).

⁷Due to the time convention $\exp(j\omega t)$ it is in fact the negative imaginary part that is of interest, and therefore the minimum of the curve.

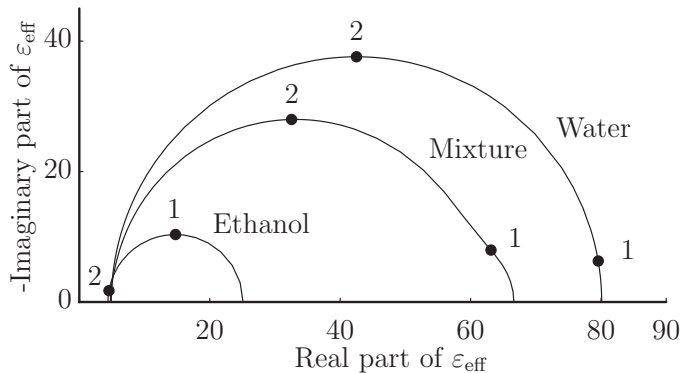


Figure 5: The Cole–Cole plot of the mixture of spherical ethanol droplets in water (20% ethanol). Shown are also the Cole–Cole diagrams of pure water and pure ethanol. Two frequency points are marked on each curve: 1 – the relaxation frequency of ethanol (1.33 GHz) and 2 – the relaxation frequency of water (15.8 GHz). Note that the negative part of the imaginary part is given, due to the time convention $\exp(j\omega t)$.

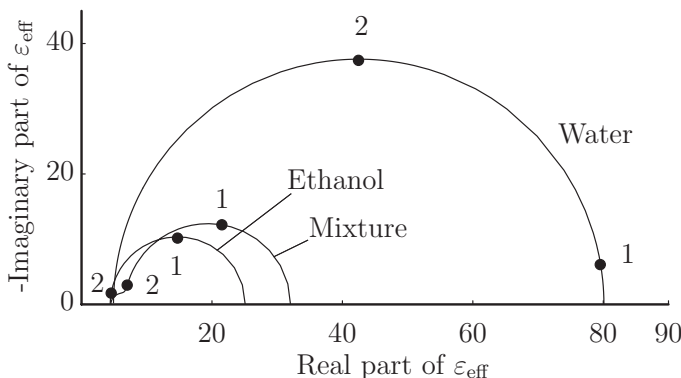


Figure 6: The same as figure 5, for a mixture of water drops in ethanol (80% ethanol).

From the figures we can observe the natural result that the mixture curve lies closer to the curve of the background medium, being composed of four fifths of it. But more interesting is certainly the frequency behavior of the mixture permittivity. It is not a Debye-type semi-circle but there exists a bend in the curve, which is more conspicuous at low frequencies for the alcohol–in–water mixture and at high frequencies for the water–in–alcohol mixture. This bend is reminiscent of the curve shape of a Davidson–Cole formula for the frequency dependence of the permittivity of polar materials [17, Sec. 3.7].

The time domain analysis of the same Debye–in–Debye mixture is given later in Section 5.2.3.

5.2 Time domain

Moving again back into the time domain, the mixing analysis can also be performed using permittivity operators as discussed in Section 3.2. In this case the multiplications and divisions with permittivity parameters have to be replaced by convolution operators and inversions of such operators. To illustrate the analysis, let us consider a mixture with dispersive inclusions in dispersionless background. The time domain Maxwell Garnett formula, (3.6), written for a non-dispersive background medium, $\epsilon_b = \epsilon_b \delta$, and dispersive inclusions with optical response ϵ_∞ and susceptibility kernel $\chi(t)$, whence $\epsilon_i = \epsilon_\infty + \chi*$, looks like

$$\epsilon_{\text{eff}} = \epsilon_b + 3\epsilon_b f(\epsilon_\infty - \epsilon_b + \chi*)[\epsilon_\infty + 2\epsilon_b - f(\epsilon_\infty - \epsilon_b) + (1 - f)\chi*]^{-1} \quad (5.10)$$

This expression is well defined unless $\epsilon_\infty + 2\epsilon_b - f(\epsilon_\infty - \epsilon_b) = 0$. For several types of susceptibility kernels, the required integrations for the operator products and inverses can be carried out analytically. We here discuss the inverse calculations in more detail and give as examples the inverses for certain material models.

5.2.1 Debye model

For example, the susceptibility kernel in the Debye model is

$$\chi(t) = H(t)\beta e^{-t/\tau} \quad (5.11)$$

where $H(t)$ is the Heaviside step function⁸ to guarantee causality, and the constant β is connected to the parameters in (5.1) by $\beta\tau = \epsilon_s - \epsilon_\infty$. The time- and frequency domain descriptions form a Fourier-transform pair (2.2) as can be checked by comparing (3.3), (5.1), and (5.11).

In the evaluation of ϵ_{eff} according to (5.10), the following results are needed. For two exponential functions $\chi_i(t) = H(t)\beta_i e^{-t/\tau_i}$ (where $i = 1, 2$) the convolution is

$$(\chi_1 * \chi_2)(t) = H(t)\beta_1\beta_2 \frac{e^{-t/\tau_1} - e^{-t/\tau_2}}{1/\tau_2 - 1/\tau_1}$$

In case the relaxation times are equal ($\tau_1 = \tau_2 = \tau$) this expression has to be replaced by

$$(\chi_1 * \chi_2)(t) = H(t)\beta_1\beta_2 t e^{-t/\tau}$$

Furthermore, the resolvent kernel of (5.11) is

$$\chi_{\text{res}}(t) = -H(t)\beta e^{-(1+\beta\tau)t/\tau}$$

Using these results, the effective permittivity operator (5.10) can be written for the Debye-mixture (inclusions of the form (5.11)) after some algebra:

$$\epsilon_{\text{eff}} = \epsilon_{\infty, \text{eff}} + \chi_{\text{eff}}(t)* \quad (5.12)$$

⁸The Heaviside step function is zero for negative arguments, and one for positive ones.

| Model | $\chi(t)$ | $\varepsilon(\omega) - \varepsilon_\infty$ | Note |
|----------------|---|---|------------------------------------|
| Debye | $\beta e^{-t/\tau}$ | $\frac{\beta\tau}{1 + j\omega\tau}$ | |
| Lorentz | $\frac{\omega_p^2}{\nu_0} \sin(\nu_0 t) e^{-\nu t/2}$ | $\frac{\omega_p^2}{\omega_0^2 - \omega^2 + j\omega\nu}$ | $\nu_0^2 = \omega_0^2 - (\nu/2)^2$ |
| Drude | $\frac{\omega_p^2}{\nu} (1 - e^{-\nu t})$ | $\frac{\omega_p^2}{-\omega^2 + j\omega\nu}$ | |
| Modified Debye | $\omega_p^2 t e^{-\nu t/2}$ | $\frac{\omega_p^2}{(\nu/2 + j\omega)^2}$ | |

Table 1: Susceptibility kernels $\chi(t)$ and the corresponding frequency-dependent permittivity functions for the models analyzed in Section 5.

with

$$\varepsilon_{\infty, \text{eff}} = \varepsilon_b + 3\varepsilon_b f \frac{\varepsilon_\infty - \varepsilon_b}{\varepsilon_\infty + 2\varepsilon_b - f(\varepsilon_\infty - \varepsilon_b)}$$

and

$$\chi_{\text{eff}}(t) = \frac{9f\varepsilon_b^2\beta H(t)}{[\varepsilon_\infty + 2\varepsilon_b - f(\varepsilon_\infty - \varepsilon_b)]^2} \exp\left[-\left(\frac{1}{\tau} + \frac{(1-f)\beta}{\varepsilon_\infty + 2\varepsilon_b - f(\varepsilon_\infty - \varepsilon_b)}\right)t\right] \quad (5.13)$$

This result shows the time domain correspondence of the MG result in frequency domain. For example, the decrease of the relaxation time constant is evident from (5.13), and agrees with the earlier result which was illustrated in Figure 2. In fact, the effective relaxation time constant in (5.13) is easily proved to be identical to the result in (5.2).

5.2.2 Other models

The above example with the Debye model showed that knowing the resolvent kernel of the basic material response is essential in the time domain calculation of the effective permittivity. For assistance in time domain mixture analysis, we can write the following two tables for the basic models that have been treated above in the frequency domain. Table 1 gives the frequency domain permittivity function for each susceptibility kernel and Table 2 shows the resolvents.⁹ All kernels are defined only for non-negative values (a Heaviside step function is assumed and omitted).

Note that the Drude model is a special case of the Lorentz model, and can be obtained as the limit $\omega_0 \rightarrow 0$ (meaning $\nu_0 = j\nu/2$). The modified Debye model is also a special case of the Lorentz model, and can be written from it as the limit $\nu_0 \rightarrow 0$ (meaning $\omega_0 = \nu/2$).

⁹The resolvent kernels can be used in calculating operators not only for mixing formulas but also for many other time domain quantities.

| Model | $\chi(t)$ | $\chi_{\text{res}}(t)$ | Note |
|----------------|---|--|---------------------------------------|
| Debye | $\beta e^{-t/\tau}$ | $-\beta e^{-(1/\tau+\beta)t}$ | |
| Lorentz | $\frac{\omega_p^2}{\nu_0} \sin(\nu_0 t) e^{-\nu t/2}$ | $-\frac{\omega_p^2}{\omega_r} \sin(\omega_r t) e^{-\nu t/2}$ | $\omega_r^2 = \nu_0^2 + \omega_p^2$ |
| Drude | $\frac{\omega_p^2}{\nu} (1 - e^{-\nu t})$ | $-\frac{\omega_p^2}{\omega_r} \sin(\omega_r t) e^{-\nu t/2}$ | $\omega_r^2 = \omega_p^2 - (\nu/2)^2$ |
| Modified Debye | $\omega_p^2 t e^{-\nu t/2}$ | $-\omega_p \sin(\omega_p t) e^{-\nu t/2}$ | |

Table 2: Susceptibility kernels $\chi(t)$ and the corresponding resolvent kernels $\chi_{\text{res}}(t)$ for the models analyzed in Section 5. The Heaviside step function $H(t)$ has been omitted.

5.2.3 A mixture of two dispersive materials

If the background medium and the inclusions are both dispersive, the time domain MG formula can still be written in the form, (5.12), where the optical response of the mixture, $\varepsilon_{\infty, \text{eff}}$, is given in (5.9) and the effective susceptibility kernel, $\chi_{\text{eff}}(t)$, is

$$\chi_{\text{eff}}(t) = \chi_b(t) + A(t) + (\varepsilon_{\infty, \text{eff}} - \varepsilon_{\infty, b}) B_{\text{res}}(t) + (A * B_{\text{res}})(t) \quad (5.14)$$

The kernels $A(t)$ and $B(t)$ are found to be

$$\begin{cases} A(t) = (\varepsilon_{\infty, \text{eff}} - \varepsilon_{\infty, b}) \left(\frac{\chi_b(t)}{\varepsilon_{\infty, b}} + \frac{\chi_i(t) - \chi_b(t)}{\varepsilon_{\infty, i} - \varepsilon_{\infty, b}} + \left(\frac{\chi_b}{\varepsilon_{\infty, b}} * \left(\frac{\chi_i - \chi_b}{\varepsilon_{\infty, i} - \varepsilon_{\infty, b}} \right) \right) (t) \right) \\ B(t) = f_b \chi_b(t) + f_i \chi_i(t) \end{cases}$$

where

$$f_b = \frac{(2 + f)}{(1 - f)\varepsilon_{\infty, i} + (2 + f)\varepsilon_{\infty, b}}, \quad f_i = \frac{(1 - f)}{(1 - f)\varepsilon_{\infty, i} + (2 + f)\varepsilon_{\infty, b}}$$

Thus, the effective susceptibility kernel can be obtained by solving a resolvent equation followed by straightforward convolution. The resolvent kernel $B_{\text{res}}(t)$ cannot be written in closed form in general.

We now focus on the special case of a Debye-in-Debye mixture which was analyzed in frequency domain in Section 5.1.5:

$$\begin{cases} \chi_b(t) = \frac{\varepsilon_{s, b} - \varepsilon_{\infty, b}}{\tau_b} \exp\left(-\frac{t}{\tau_b}\right) H(t) \equiv \alpha_b \exp(-t\beta_b) H(t) \\ \chi_i(t) = \frac{\varepsilon_{s, i} - \varepsilon_{\infty, i}}{\tau_i} \exp\left(-\frac{t}{\tau_i}\right) H(t) \equiv \alpha_i \exp(-t\beta_i) H(t) \end{cases}$$

In the case $\tau_b \neq \tau_i$,

$$A(t) = a_1 \chi_b(t) + a_2 \chi_i(t) + a_3 \chi_b(t) \frac{t}{\tau_b}$$

where

$$\begin{cases} a_1 = \frac{\varepsilon_{\infty,\text{eff}} - \varepsilon_{\infty,b}}{\varepsilon_{\infty,b}} - \frac{\varepsilon_{\infty,\text{eff}} - \varepsilon_{\infty,b}}{\varepsilon_{\infty,i} - \varepsilon_{\infty,b}} \left(1 + \frac{\varepsilon_{s,i} - \varepsilon_{\infty,i}}{\varepsilon_{\infty,b}} \frac{\tau_b}{\tau_i - \tau_b} \right) \\ a_2 = \frac{\varepsilon_{\infty,\text{eff}} - \varepsilon_{\infty,b}}{\varepsilon_{\infty,i} - \varepsilon_{\infty,b}} \left(1 + \frac{\varepsilon_{s,b} - \varepsilon_{\infty,b}}{\varepsilon_{\infty,b}} \frac{\tau_i}{\tau_i - \tau_b} \right) \\ a_3 = -\frac{(\varepsilon_{\infty,\text{eff}} - \varepsilon_{\infty,b})(\varepsilon_{s,b} - \varepsilon_{\infty,b})}{\varepsilon_{\infty,b}(\varepsilon_{\infty,i} - \varepsilon_{\infty,b})} \end{cases}$$

and

$$B_{\text{res}}(t) = -\chi_+(t) - \chi_-(t) \equiv -\alpha_+ \exp(-\beta_+ t)H(t) - \alpha_- \exp(-\beta_- t)H(t)$$

where

$$\begin{cases} 2\beta_{\pm} = f_i \alpha_i + \beta_i + f_b \alpha_b + \beta_b \pm \sqrt{(f_i \alpha_i - \beta_i + f_b \alpha_b + \beta_b)^2 + 4f_i \alpha_i (\beta_i - \beta_b)} \\ \alpha_{\pm} = \frac{\beta_i - \beta_b}{\frac{(\beta_{\mp} - \beta_b)}{(\beta_{\pm} - \beta_b)} - \frac{(\beta_{\mp} - \beta_i)}{(\beta_{\pm} - \beta_i)}} \end{cases}$$

Using (5.14) gives the explicit expression for the effective susceptibility kernel:

$$\chi_{\text{eff}}(t) = c_1 \chi_b(t) + c_2 \chi_i(t) + c_3 \chi_+(t) + c_4 \chi_-(t) + c_5 \chi_b(t) \frac{t}{\tau_b}$$

where the coefficients

$$\begin{cases} c_1 = 1 + a_1 \left(1 - \frac{\alpha_+}{\beta_+ - \beta_b} - \frac{\alpha_-}{\beta_- - \beta_b} \right) + a_3 \left(\frac{\alpha_+ \beta_b}{(\beta_+ - \beta_b)^2} + \frac{\alpha_- \beta_b}{(\beta_- - \beta_b)^2} \right) \\ c_2 = a_2 \left(1 - \frac{\alpha_+}{\beta_+ - \beta_i} - \frac{\alpha_-}{\beta_- - \beta_i} \right) \\ c_3 = -\varepsilon_{\infty,\text{eff}} + \varepsilon_{\infty,b} - a_1 \frac{\alpha_b}{\beta_b - \beta_+} - a_2 \frac{\alpha_i}{\beta_i - \beta_+} - a_3 \frac{\alpha_b \beta_b}{(\beta_+ - \beta_b)^2} \\ c_4 = -\varepsilon_{\infty,\text{eff}} + \varepsilon_{\infty,b} - a_1 \frac{\alpha_b}{\beta_b - \beta_-} - a_2 \frac{\alpha_i}{\beta_i - \beta_-} - a_3 \frac{\alpha_b \beta_b}{(\beta_- - \beta_b)^2} \\ c_5 = a_3 \left(1 - \frac{\alpha_+}{\beta_+ - \beta_b} - \frac{\alpha_-}{\beta_- - \beta_b} \right) \end{cases}$$

are real and dimensionless. Consequently, $\chi_{\text{eff}}(t)$ is a linear combination of four Debye kernels and one modified Debye kernel. It can be shown that

$$\min(\beta_i, \beta_b) < \beta_- < \max(\beta_i, \beta_b) < \beta_+, \quad \alpha_{\pm} > 0$$

This implies that the relaxation time $\tau_+ = 1/\beta_+$ is less than both the relaxation times τ_i and τ_b , whereas $\tau_- = 1/\beta_-$ assumes a value between τ_i and τ_b .

Numerical results for a Debye-in-Debye medium (ethanol and water, 20-80% and 80-20%) using equation (5.14) are displayed in Figure 7. The figure shows that although the mixture curves resemble in form the Debye-kernels of water and ethanol, they are not exactly exponentially decaying. The differences correspond

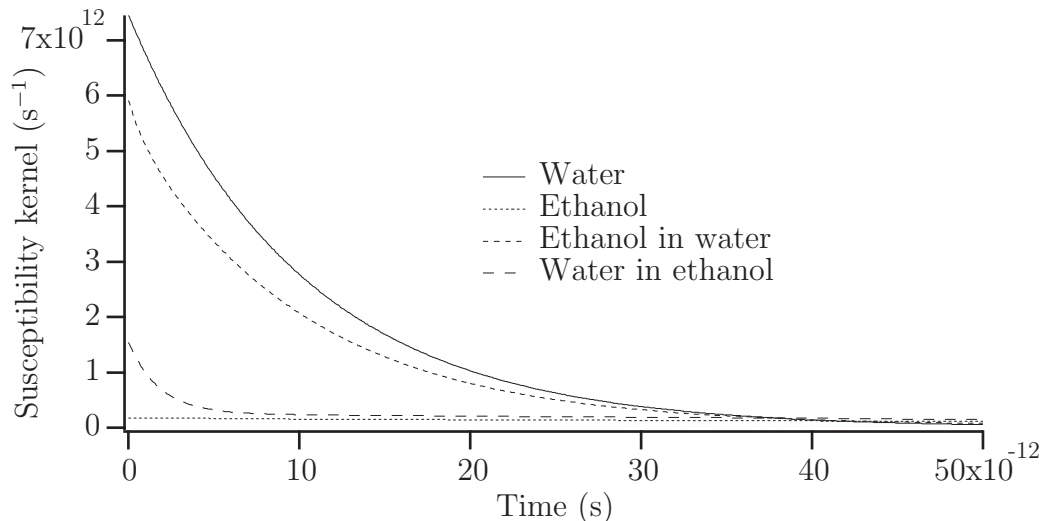


Figure 7: The susceptibility kernels for water, ethanol, ethanol-in-water mixture (20% ethanol), and water-in-ethanol mixture (80% ethanol).

to the bends of the Cole–Cole curves that were observed in Figures 5 and 6. It is worth observing from the curves that as far as the early-time responses (t small) are concerned, both mixtures resemble water which can be seen from the quick decay of the kernel. This is different from the stable early-time response of ethanol. Another interesting detail of Figure 7 is the fact that the water-in-ethanol mixture has a slightly stronger memory for times after 40 ps compared to both water and ethanol, although the difference is very small.

6 Discussion and conclusions

The foregoing analysis has hopefully cast light upon the two different approaches of the homogenization problem of mixtures. The classical treatment of permittivities as frequency-dependent quantities takes the plain static Maxwell Garnett rule and allows the parameters to vary with frequency. With this approach it is easy to calculate the frequency behavior of the macroscopic permittivity of a mixture.

However, if one desires to find out the temporal behavior of the susceptibility kernel of a mixture, the frequency domain approach does not always work directly. It is true, as was shown in Section 5, that for certain dispersion models for the inclusion permittivity the mixture permittivity can also be observed to follow a known model which then allows one to identify the susceptibility kernel in the time domain. But in the general case one needs to perform a Fourier inversion of the mixture permittivity $\varepsilon_{\text{eff}}(\omega)$ to find out the kernel.

A direct time domain approach that has been developed in the present paper expresses the effective permittivity of a mixture as an operator-form Maxwell Garnett expression which includes the susceptibility kernels and optical responses of the component materials. The evaluation of the effective permittivity operator requires

calculation of convolutions and operator inverses. The inverse of the convolution operators encountered in these problems can be solved from Volterra equations of the second kind which have unique and well-behaving solutions. The advantage of the time domain formulation is that the results are closer to physics than the frequency domain results. As the example with alcohol–water mixture showed, the features of the time domain result (Figure 7) were easier to interpret than the characteristics of Figures 5 and 6.

In terms of geometry of the mixture, the present analysis is limited to the simplest case where the inclusions are spherical. The ellipsoidal form is another one suited for straightforward polarizability analysis because the internal static field in ellipsoids, too, is also uniform. Then the effective permittivity functions contain the depolarization factors of the ellipsoids [14]. The depolarization factors have accordingly an effect on the manner how the dispersion characteristics of the mixture differ from those of the inclusions and the background. Indeed, the frequency dependence of a mixture permittivity could be used as an indicator of the microstructure of heterogeneous media that contain strongly dispersive components. An example could be wet snow where the frequency dependence of liquid water determines the dispersion of snow permittivity. The pendular–funicular transition in wet snow around a few per cent wetness point is an experimentally known fact [7], and this transition certainly has a connection to the shape the water droplets occupy in the lattice of ice grains.

One has to remember the quasi-static limitation when making use of mixing rules. The analysis of the present paper assumed that the internal field of an inclusion is spatially uniform. This means that the inclusions have to be small compared to the distance which electromagnetic wave propagates during a time constant of the temporal variation of the applied field. One has also to be careful with conducting materials for which the field amplitude suffers a decay inwards from the inclusion surface. It is important to check that the penetration depth is greater or at least of the order of the sphere diameter. As an example take the case of clouds as a water-droplet mixture which was analyzed in Section 5.1.1. The penetration depth is $c/\left[\omega\text{Im}\sqrt{\varepsilon(\omega)}\right]$, which is around $600\ \mu\text{m}$ at the frequency of 30 GHz, and around $200\ \mu\text{m}$ at 300 GHz. These figures assure that certainly all field inhomogeneities caused by finite penetration depth can be ignored for those droplet sizes that can be present in fog and clouds.

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Appendix A Inverse operator and resolvent

The calculation of an inverse for convolution operators makes use of the so-called resolvent operator. Consider first a permittivity-type operator of the form $\epsilon = 1 + \chi^*$ where we first assume for simplicity the optical response to be one. The inverse to this operator contains the resolvent operator which is also an integral operator:

$$\epsilon^{-1} = (1 + \chi^*)^{-1} \equiv 1 + \chi_{\text{res}}^* = (\delta + \chi_{\text{res}})^*$$

This operator satisfies $\epsilon^{-1}\epsilon = \epsilon\epsilon^{-1} = 1$.

The resolvent kernel of $\chi(t)$ is denoted by $\chi_{\text{res}}(t)$ and satisfies the linear Volterra integral equation of the second kind

$$\chi_{\text{res}}(t) + \chi(t) + (\chi_{\text{res}} * \chi)(t) = 0$$

This equation has a unique solution in the space of bounded and smooth functions in each bounded time-interval $0 < t < T$, see, *e.g.*, [12, p. 33]. Explicitly, the resolvent kernel can be represented by the function series¹⁰

$$\chi_{\text{res}}(t) = \sum_{k=1}^{\infty} (-1)^k ((\chi^*)^{k-1} \chi)(t)$$

The resolvent kernel vanishes for $t < 0$ and has a finite jump-discontinuity at $t = 0$ if and only if $\chi(t)$ has such a discontinuity: $\chi_{\text{res}}(0+) = -\chi(0+)$.

In the mixing calculations we need inverses of operators where the optical response is different than one, see, *e.g.*, (3.4). A similar deconvolution procedure gives us the inverse: let the inverse of an operator $a + A^*$ be written as $b + B^*$, where a and b are nonzero constants. Then we have

$$ab + [aB + bA + A * B]^* = 1 = \delta^*$$

which means that $b = 1/a$ and the unknown kernel $B(t)$ can be solved from the resolvent equation

$$aB(t) + bA(t) + (A * B)(t) = 0$$

This is again a Volterra equation of the second kind for B , which is a well-behaving problem numerically.

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¹⁰This solution is known as the method of successive approximations.

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