Modeling of Isotropic Backward-Wave Materials Composed of Resonant Spheres

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Abstract

A possibility to realize isotropic artificial backward-wave materials is theoretically analyzed. An improved mixing rule for the effective permittivity of a composite material consisting of two sets of resonant dielectric spheres in a homogeneous background is presented. The equations are validated using the Mie theory and numerical simulations. The effect of a statistical distribution of sphere sizes on the increasing of losses in the operating frequency band is discussed and some examples are shown.

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I. INTRODUCTION

Most of the known realizations of artificial backward-wave materials with negative parameters (also called Veselago media, negative index materials, and double-negative materials) [1, 2] are highly anisotropic composites. The usual design utilizes arrays of thin conductors and arrays of split rings. In addition to anisotropy and, in most cases, even bi-anisotropy of these structures, they exhibit strong spatial dispersion due to the presence of long conductors that support TEM modes [3]. However, for many applications, isotropic materials are required or preferable. To provide isotropic response with negligible spatial dispersion, one can possibly use regular or random arrays of resonant particles that have both electric and magnetic resonances. One possibility is to use Ω-shaped metal inclusions. This opportunity has been explored theoretically in [4, 5] and experimentally in [6]. The use of racemic mixtures of chiral particles is also a possibility [7]. However, in both these solutions the inclusions are bi-anisotropic particles, and the material behaves as an effectively isotropic medium only at the macroscopic scale, if there are enough particles distributed in a proper fashion. This means that the use of isotropic particles is clearly preferable.

The idea of an isotropic backward-wave material constructed from small isotropic spheres in a dielectric background was presented in [8]. In that paper, the use of two sets of spheres was proposed. In one set, the spheres are made of a high-permittivity dielectric, and in the other set they are made of a high-permeability magnetic material. Here, resonant dielectric spheres provide effective negative permittivity, and resonant magnetic spheres provide negative permeability. In paper [9], a more practical design was suggested, such that all spheres are made of a dielectric material, but there are two sets of spheres with different radii. The dielectric constant of spheres is larger than that of the background. Then the wavelength inside the sphere is comparable to the diameter of the sphere and simultaneously the wavelength outside the sphere is large compared to the sphere. By combining two sets of spheres with suitable radii, one set of spheres is in the magnetic resonance mode and the other set is in the electric resonance mode.

The effective permittivity and permeability can be calculated using effective medium models if the wavelength outside spheres is large compared to sphere diameters and the polarizability of spheres is known. An illustration of the homogenization problem is shown in Fig. 1. Earlier studies [8, 9] are based on effective medium theories for a composite
material consisting of spheres which resonate either in the electric or magnetic mode, as presented in [10]. In Lewin’s model, spheres are assumed to resonate either in the first or second resonance mode of the Mie theory. In [8, 9] Lewin’s equations were applied directly to the system of two sets of spheres, and the electric polarizability of spheres in the magnetic resonant mode was not taken into account. However, electrical properties of these spheres can have a significant effect on the effective permittivity of the composite. This affects especially the low frequency limit, which then approaches to the classical Maxwell-Garnett mixing rule. In this paper we present an effective medium model, which takes this effect into account. Equations are validated both analytically and numerically. Scattering from a single sphere is calculated both analytically from the full Mie theory and numerically. It is shown numerically, that there exists a backward wave in the frequency band, where the effective medium theory predicts it.

The electric and magnetic polarizabilities of spheres can be calculated using the Mie theory [11]. The scattered electromagnetic field is expressed as an infinite series of vector spherical harmonics $M_n$ and $N_n$, each weighted by appropriate amplitude coefficients $a_n$ and $b_n$. These represent the normal electromagnetic modes of the spherical particle. In general, the field is a superposition of normal modes of fields. This can be interpreted as a superposition of fields generated by electric and magnetic dipoles, and multipoles. When the permittivity of the spheres is much larger than the permittivity of the environment, around the eigenfrequencies of the first two modes $a_1$ and $b_1$, the dipole fields dominate. Therefore frequency dependent polarizabilities can be introduced: The problem is still quasistatic, because the field outside the sphere can be modeled with a dipole field. For each order $n$ there are two distinct types of modes: transverse magnetic (TM) and transverse electric (TE) modes. When $n = 1$, these modes represent the field scattered by a magnetic dipole (TM) and by an electric dipole (TE). In the low frequency limit, these frequency dependent polarizabilities approach static polarizabilities.

The statistical size distribution of spheres, which is the actual case when this material is realized, is also analyzed. The size distribution causes increasing losses in the frequency band, where backward waves can exist.
II. MIXING THEORY

In this section, the Clausius-Mossotti (Maxwell-Garnett) mixing relation and polarizabilities of spheres near the first two Mie resonance modes are presented. The material of spheres and the background are assumed to be dielectric and non-magnetic. Footnote e corresponds to spheres in the electric resonance mode and m to the magnetic resonance mode.

The effective permittivity $\varepsilon_{\text{eff}}$ for a material with two types of inclusions having two different electric polarizabilities can be calculated from the generalized Clausius Mossotti relation [12]:

\[
\frac{\varepsilon_{\text{eff}} - \varepsilon_b}{\varepsilon_{\text{eff}} + 2\varepsilon_b} = \frac{n_e\alpha_e}{3\varepsilon_b} + \frac{n_m\alpha_m}{3\varepsilon_b}
\]

where $n_m$ and $n_e$ are the number of spheres per unit volume in the magnetic resonance and in the electric resonance, respectively, $\alpha_m$ and $\alpha_e$ are the electric polarizabilities of spheres in the magnetic resonance and in the electric resonance mode. In [8, 9] it was assumed that $\alpha_m = 0$. Neglecting $\alpha_m$ leads to an error which is usually small near the resonant frequency, but the error out from the resonant frequency can be quite significant. However, the error near the resonant frequency increases as the electrical contrast between inclusions and the environment increases. Also the low frequency limit for the effective permittivity is incorrect, because the remaining static electric polarizability of spheres in the magnetic resonance modes is not taken into account. The effective permeability can be obtained from (1) by replacing $\varepsilon$ with $\mu$ and the electric polarizability $\alpha$ with the magnetic polarizability $\beta$. We can assume that the magnetic polarizability of spheres in the electric resonance $\beta_e = 0$, because materials are non-magnetic: $\mu_b = \mu_i = 1$. Then the magnetic polarizability arises only from resonant phenomena, and there is no remaining static value. Therefore, the effective permeability $\mu_{\text{eff}}$ reads:

\[
\frac{\mu_{\text{eff}} - 1}{\mu_{\text{eff}} + 2} = \frac{n_m\beta_m}{3\mu_b}
\]

where $\beta_m$ is the magnetic polarizability of spheres in the magnetic resonance mode.

Frequency dependent polarizabilities $\alpha$ and $\beta$ for a sphere with radius $r$ are calculated using the Mie theory. Polarizability $\alpha_e$ corresponds to the electric polarizability with $r = r_e$, $\alpha_m$ to that with $r = r_m$, and $\beta_m$ corresponds to the magnetic polarizability with $r = r_m$. The coefficients for the spherical harmonics in the first two Mie resonance modes (where
\[ n = 1 \text{) are } [10, 11]: \]

\[ a_1 = j \frac{2}{3} (k_0^2 \mu_b \epsilon_b)^{3/2} \frac{\mu_b - \mu_i F(\Theta)}{2 \mu_b + \mu_i F(\Theta)} r^3 \]

\[ b_1 = j \frac{2}{3} (k_0^2 \mu_b \epsilon_b)^{3/2} \frac{\epsilon_b - \epsilon_i F(\Theta)}{2 \epsilon_b + \epsilon_i F(\Theta)} r^3 \]

where

\[ F(\Theta) = \frac{2(\sin \Theta - \Theta \cos \Theta)}{(\Theta^2 - 1) \sin \Theta + \Theta \cos \Theta}, \]

\[ \Theta = k_0 r \sqrt{\epsilon_i \mu_i} \]

The scattering parameter \( S_1 \) [11] for a sphere, when only the order \( n = 1 \) modes are excited reads:

\[ S_1 = \frac{3}{2} (a_1 + b_1) \]

This corresponds to scattering from electric and magnetic dipoles: \( S_1 = S_{1e} + S_{1m} \). Scattering parameter \( S_{1e} \) for an electric dipole can be written in terms of the electric polarizability \( \alpha \):

\[ S_{1e} = \frac{j k^3 \alpha}{4 \pi \epsilon_b} \]

The scattering parameter for a magnetic dipole is

\[ S_{1m} = \frac{j k^3 \beta}{4 \pi \mu_b} \]

By combining equations (3–9) with \( \mu_b = 1 \) and \( V_{\text{sphere}} = 4 \pi r^3 / 3 = f / n \), where \( f \) is the volume fraction of spheres \( f = V_{\text{sphere}} / V_{\text{tot}} \), the polarizabilities of electric and magnetic dipoles can be written as:

\[ \alpha = 4 \pi r^3 \epsilon_b \frac{\epsilon_b - \epsilon_i F(\Theta)}{2 \epsilon_b + \epsilon_i F(\Theta)} = \frac{3f \epsilon_b \epsilon_b - \epsilon_i F(\Theta)}{n} \]

\[ \beta = 4 \pi r^3 \frac{1 - \mu_i F(\Theta)}{2 + \mu_i F(\Theta)} = \frac{3f \frac{1 - \mu_i F(\Theta)}{n}}{2 + \mu_i F(\Theta)} \]

When these are substituted in the Clausius-Mossotti relations (1–2) with \( r = r_e \) for spheres in the electric resonance and \( r = r_m \) for spheres in the magnetic resonance, effective medium models for a composite consisting of two set of resonating spheres read:

\[ \frac{\epsilon_{\text{eff}} - \epsilon_b}{\epsilon_{\text{eff}} + 2 \epsilon_b} = f_e \left( \frac{2 \epsilon_b + \epsilon_i F(\Theta_e)}{\epsilon_b - \epsilon_i F(\Theta_e)} \right) + f_m \left( \frac{2 \epsilon_b + \epsilon_i F(\Theta_m)}{\epsilon_b - \epsilon_i F(\Theta_m)} \right) \]

\[ \frac{\mu_{\text{eff}} - 1}{\mu_{\text{eff}} + 2} = f_m \left( \frac{2 + F(\Theta_m)}{1 - F(\Theta_m)} \right) \]
where $\Theta_e = k_0r_e\sqrt{\varepsilon_i\mu_i}$ and $\Theta_m = k_0r_m\sqrt{\varepsilon_i\mu_i}$. These equations (12, 13) are similar to those given in [8, 9] if the second term in the right hand side in (12) is set to zero.

In Fig. 2 an example of effective permittivity as a function of the volume fraction of spheres is shown. The solid line represents $\epsilon_{eff}$ given by Eq. (12), and the dashed line $\epsilon_{eff}$ is calculated using the method of [8, 9], where the electric polarizability of spheres in the magnetic resonance mode is not taken into account. It can be seen that the resonant frequency slightly shifts when the improved mixing equation is used. In this case the old effective medium model overestimates the effective permittivity out of the resonance mode.

III. STATISTICAL SIZE DISTRIBUTION OF SPHERES

In practice, when a set of spheres is manufactured, the sphere dimensions are statistically distributed because of production inaccuracies. The effect of a continuous statistical size distribution on the real part of the effective permittivity and the losses will be estimated next. The Clausius-Mossotti equation for $K$ spheres in electric resonance and $N$ spheres in magnetic resonance reads:

$$\frac{\epsilon_{eff} - \epsilon_b}{\epsilon_{eff} + 2\epsilon_b} = \frac{1}{3\epsilon_b} \left( \sum_{k=1}^{K} n_{ek}\alpha_{ek} + \sum_{n=1}^{N} n_{mn}\alpha_{mn} \right)$$

(14)

By substituting the electric polarizabilites (10) into equation (14), with $r = r_e$ for the spheres in the electric resonance and $r = r_m$ for the spheres in the magnetic resonance, we get:

$$\frac{\epsilon_{eff} - \epsilon_b}{\epsilon_{eff} + 2\epsilon_b} = \sum_{k=1}^{K} \frac{f_{ek}}{G(\Theta_{ek})} + \sum_{n=1}^{N} \frac{f_{mn}}{G(\Theta_{mn})}$$

(15)

This is a solution for the effective permittivity with two sets of spheres. One set of spheres is in the magnetic resonance and the other set is in the electric resonance. The effective permeability can be calculated in a similar way.

Eq. (15) is a discrete summation. It can be interpreted also as samples taken with the Dirac delta function from continuous probability density functions $g_1(r_e)$ and $g_2(r_m)$, which describe the particle size distributions of spheres in the electric and magnetic resonances. They are normalized as $\int_0^\infty g_1(r_e) \, dr_e = \int_0^\infty g_2(r_m) \, dr_m = 1$.

Then the effective permittivity for the continuous size distributions reads:

$$\frac{\epsilon_{eff} - \epsilon_b}{\epsilon_{eff} + 2\epsilon_b} = f_e \int_0^\infty \frac{g_1(r_e)}{G(\Theta_e)} \, dr_e + f_m \int_0^\infty \frac{g_2(r_m)}{G(\Theta_m)} \, dr_m$$

(16)
Here
\[
G(\Theta_i) = \frac{\epsilon_b - \epsilon_i F(\Theta_i)}{2\epsilon_b + \epsilon_i F(\Theta_i)}
\] (17)

functions \(1/G(\Theta_e)\) and \(1/G(\Theta_m)\) are integrable, \(f_e\) is the volume fraction of spheres in the electric resonance, and \(f_m\) is the volume fraction of spheres in the magnetic resonance mode.

An example on how a normal size distribution \(N = 1/(\sigma \sqrt{2\pi}) \exp((r - \hat{r})^2/2)\) of spheres with half value widths \(\sigma = \sigma_e\) and \(\sigma = \sigma_m\) and expectation values \(\hat{r} = r_e\) and \(\hat{r} = r_m\) affects the effective material parameters is presented in Figs. 3(a) and 3(b). The half-value widths \(\sigma_e = \sigma_m = 1 \mu m\) (Fig. 3(a)) does not increase losses significantly, but with the half-value widths of 10 \(\mu m\) (Fig. 3(b)) the imaginary part becomes large and \(\mu_{eff}\) does not take negative values.

We have not analyzed how the nonidealities in the lattice structure will affect the scattering losses or how scattering losses will increase because of the size distribution of spheres.

\section*{IV. VERIFICATION OF THE EFFECTIVE MEDIUM MODEL}

In the original Lewin’s paper [10], the validity limit of effective medium equations was given as \(\lambda/|\sqrt{\epsilon_{eff}\mu_{eff}}| > 10r\). This limit is very strict, because it was derived by demanding reasonable values when the filling fraction \(f \rightarrow \infty\) in the Clausius-Mossotti mixing equation. This requirement is not necessary, because the mixing equation is anyway not valid when \(f \rightarrow \infty\).

In this paper, the frequency dependent polarizabilities for spheres are given. To verify when these polarizabilities are valid, one should study the complete Mie theory expansion, where all the higher order modes are taken into account. If the spheres can be replaced with frequency dependent electric and magnetic dipoles (10, 11), then also the Clausius-Mossotti effective medium models are valid when the volume fraction of spheres is small. To study how well the effective medium model works for high volume fractions of spheres, one should first study if close separation of spheres causes excitation of higher order resonance modes.

There are two limitations in the model. The first limitation is that the wavelength outside the spheres should be large compared to the size of spheres, so that the material would work as an effective medium and the spheres could be modeled by electric and magnetic dipoles with frequency dependent polarizabilities. The second limitation arises from the mixing theory: the filling fraction of spheres should be small.
The effective medium model is based on the assumption, that the spheres resonate in the first (magnetic) and the second (electric) resonance modes. This assumption can be tested using the Mie theory. In Fig. 4(a) the scattering cross section \( Q_{\text{sca}} \) of a sphere with radii \( r = r_e = 3.18 \text{ mm} \) and \( r = r_m = 2.28 \text{ mm} \) and \( \epsilon_i = 44(1-j10^{-4}) \) is shown. Radii of spheres were chosen so that the electric resonance and magnetic resonance occur at approximately the same frequency. The smaller sphere resonates clearly in the first resonance mode (magnetic), because no additional peaks can be seen. The larger sphere has the second resonant mode (electric) near the same frequency, but the third resonant mode has its resonant frequency near the second mode. This mode is not taken into account in the effective medium models (12, 13), which are based on the first two resonant modes. However, \( Q_{\text{sca}} \) goes nearly to zero between these modes, which indicates that when the sphere is in the second resonant mode, the effect of the third resonant mode can be neglected. The third resonance mode sets an upper limit for the validity of Eq. (12) for the effective permittivity, but it also sets an upper limit for the frequency band where the effective permittivity can have negative values in practice.

The existence of a backward wave can be verified numerically by plotting the electric field inside a material layer in the frequency band where the effective medium theory predicts both material parameters to be negative. Numerical simulations were made using the finite element method based on Agilent HFSS electromagnetic modeling software. To test the accuracy of the solution, a single sphere with radius \( r = r_e = 3.18 \text{ mm} \) or \( r = r_m = 2.28 \text{ mm} \) and \( \epsilon_i = 44(1-j10^{-4}) \) was placed at the center of an air filled rectangular waveguide. The height and width of the waveguide were \( w = h = 15 \text{ mm} \), and the length \( l = 15 \text{ mm} \). Boundary conditions were ideal electric conductor (PEC) at the top and bottom of the waveguide and ideal magnetic conductor (PMC) on the sides of the waveguide. The incident electric field was vertically polarized. The scattering matrix element \( |S_{11}| \) was solved using about 37000 unknowns. The result is shown in Fig. 4(b). Similar peaks as in the analytical solution can be seen, but the numerically calculated resonant frequencies are slightly higher. When the number of unknowns was increased to 120000, then the resonant frequencies were the same as in the analytical solution. It appears that when using smaller number of unknowns, the numerical solution overestimates the resonant frequencies about 1 %. Both the numerical and analytical studies show, that spheres can be modeled using frequency dependent polarizabilities (10, 11) up to the frequency, where the third resonant
mode appears.

A material layer consisting of both sets of spheres was modeled numerically. The accuracy was set to correspond to the simulation with one sphere with 37,000 unknowns, accordingly we should expect numerical simulations to have a frequency shift of about 1% upwards. An illustration of the calculation domain is shown in Fig. 5. Two cases were studied: two and four layers of spheres. The studied waveguide corresponds to two or four infinite layers of spheres, where the nearest neighbors of spheres with radius $r_e$ are spheres with radius $r_m$ and vice versa. In the simulations, mirror planes cut spheres into four parts. These boundary conditions do not disturb the first three resonant modes of the spheres, which have the same symmetry. The material parameters and radii were the same as for simulations with a single sphere in Fig. 4(b). The results for scattering matrix elements $|S_{11}|$ and $|S_{21}|$ are shown in Fig. 6(a) and Fig. 6(b). The effective medium model (12, 13) predicts, that this medium should have negative material parameters from 9.92 GHz to 9.98 GHz. When the frequency shift caused by numerical error is taken into account, the negative material parameters should occur around 10.05 GHz. From figures it can be seen, that around this frequency there is a propagating mode. The stop band after this pass band is because of the third resonant mode of the larger spheres. If both material parameters have negative values, the wave inside the material should be a backward wave. However, it can not be seen from $S$-parameter studies if the wave is a forward or a backward wave.

The existence of a backward wave near 10.05 GHz can be verified numerically by plotting the electric field distribution inside a material layer. For the chosen harmonic time dependence the phase of the electric field component $E_y$ decreases in the positive direction of the wave vector $k$. Fig. 7 displays the phase of the electric field at 10.064 GHz inside the simulated waveguide. Black corresponds to $-\pi$ and white to $\pi$. The feeding point is on the left, so the energy propagates from left to right, but we see that the phase increases from the left to right. This clearly shows that this is a backward wave. From similar field plots it is seen, that the wave is a backward wave in the entire frequency band around 10.1 GHz. According to the $S$-parameter studies there is a propagating mode [Figs. 6(a), 6(b)]. In the numerical simulations, the center frequency is about 10.1 GHz and the bandwidth is 1%. The center frequency calculated using the effective medium theory is 9.95 GHz, which corresponds to 10.05 GHz in Fig. 6(a) and 6(b) because of the numerical overestimation of the resonant frequencies by about 1%. The bandwidth in the effective medium theory is
about 0.6%. We can conclude that the effective medium model well predicts the frequencies where the backward wave exists. It appears that the bandwidth in the numerical simulations is even larger than the prediction of the effective medium model.

V. CONCLUSIONS

In this study, the possibility to realize an isotropic material with negative real parts of both $\epsilon$ and $\mu$ using a mixture consisting of two sets of resonating dielectric spheres with different radii was considered. A corrected effective medium model for the effective permittivity was presented, which can be used also to calculate how a statistical size distribution increases the losses in the frequency band where both material parameters have negative values. The limitations of the analytical model were discussed and numerically validated.

According to numerical studies, a thin material slab supports a backward wave in the same frequency band which the effective medium model predicts. The backward-wave regime corresponds to both $\epsilon$ and $\mu$ being negative.

In the simulations, quite low contrast between the inclusions and the environment was used. The introduced effective medium theory for resonant spheres should be even more applicable when the contrast is larger. For higher frequencies, the medium starts to behave as an electromagnetic band gap structure (EBG), where backward waves can also exist. However, EBG structures are not isotropic and they can not be modeled using effective medium theories.

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FIG. 1: A composite construct with two sets of dielectric spheres in a dielectric background. If the wavelength is much larger than the radii of the spheres and the distance between them, the material can be modeled with effective permittivity $\varepsilon_{\text{eff}}$ and permeability $\mu_{\text{eff}}$.

FIG. 2: The effective permittivity $\varepsilon_1$ as a function of the frequency calculated from Eq. (12) compared to the effective permittivity $\varepsilon_2$ calculated without taking into account the electrical polarizability of spheres in the magnetic resonance. In this case the second term on the right-hand side of Eq. (12) is zero. $\varepsilon_i = 100(1 - j0.25 \cdot 10^{-4})$, $\varepsilon_b = 1$, $f_e = 0.15$, $f_m = 0.15$, $r_e = 3.18 \cdot 10^{-3}$ mm, $r_m = 2.18 \cdot 10^{-3}$ mm.
(a) $\sigma_e = \sigma_m = 1\mu m$

(b) $\sigma_e = \sigma_m = 10\mu m$

FIG. 3: The effective permittivity as a function of the frequency calculated using Eq. (16) with $\epsilon_i = 44(1 - j1.25 \cdot 10^{-4})$, $\epsilon_b = 1$ and filling ratios $f_e = 2\%$, $f_m = 14\%$. On the left-hand side spheres are normally distributed with the half-value widths $\sigma_m = \sigma_e = 10^{-5}$ and expectation values $r_e = 3.18 \text{ mm}$, $r_m = 2.28 \text{ mm}$. The size distribution $N$ for $\sigma_e$ is also shown. On the right-hand side everything is the same, expect the half value widths are $\sigma_e = \sigma_m = 10^{-6}$. 
FIG. 4: Analytically calculated RCS (a) and numerically calculated $|S_{11}|$ (b) for a sphere with $r_e = 3.18$ mm and $\epsilon_i = 44(1 - j10^{-4})$.

FIG. 5: Two and four layers of spheres with similar cross section in the $(x, y)$-plane were studied. There are four quarter of spheres in each layer, two with radius $r_1 = 2.28$ mm and two with radius $r_2 = 3.18$ mm.
FIG. 6: Numerically calculated $S$-parameters for a slab consisting of two and four layers of spheres.

(a) $S_{11}$, $r_1 = 2.28$ mm and $r_2 = 3.18$ mm

(b) $S_{21}$, $r_1 = 2.28$ mm and $r_2 = 3.18$ mm

FIG. 7: The phase of the electric field component $E_y$ at 10.064 GHz for a structure as in Figs. 6(a), 6(b) with four layers of spheres. The feeding plane is on the left, and the energy propagates from left to right. The black color corresponds to $-\pi$ and the white to $+\pi$. The phase is increasing as the distance from the source increases, which indicates that the wave is a backward wave.