A Double Negative (DNG) Composite Medium Composed of Magnetodielectric Spherical Particles Embedded in a Matrix

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***Abstract*—We study a composite medium consisting of insulating magnetodielectric spherical particles embedded in a background matrix. Using results from the literature going back as far as Lewin (1947), we show that the effective permeability and permittivity of the mixture can be simultaneously negative for wavelengths where the spherical inclusions are resonant and that the medium results in an effective "double negative (DNG) media." Materials of this type are also called negative-index materials, backward media (BW), and left-handed materials. This type of material belongs to a more general class of metamaterials. The theoretical results presented here show that composite media having much simpler structure than those recently reported in the literature can exhibit negative permeability and permittivity over significant bandwidths.**

***Index Terms*—Composite medium, double negative, left-handed, metamaterial, spherical particles.**

I. Introduction

PERMITTIVITY arises from the induced electric-dipole response of a large number of small particles [1, pp. 159–162]. Classically, these particles have been atoms or molecules, but in the past 60 years so-called artificial dielectrics have been developed whose "atoms" are small metal or dielectric objects, large compared to atomic dimensions, but still small compared to the wavelength of the electromagnetic waves acting in the "host" medium in which these inclusions are embedded [2]–[11]. In either case, the induced dipole moments are related by the electric polarizabilities of the scatterers to the electric field acting on each one. Permeability originates from the angular momentum of charge due to particle spin and orbital movement and is related to magnetic polarizabilities of the scatterers in a similar way. The effective or averaged fields $\mathbf{E}$, $\mathbf{D}$, $\mathbf{H}$, and $\mathbf{B}$ are then related to each other by the usual expressions

$$\mathbf{D} = \epsilon\mathbf{E}; \qquad \mathbf{B} = \mu\mathbf{H}$$

where ϵ and μ are related to the polarizability densities of the scatterers in space.[1] We will denote the real and imaginary parts of the relative permittivities and permeabilities by a subscript r, where $\epsilon = \epsilon_0(\epsilon_r' - i\epsilon_r'')$ and $\mu = \mu_0(\mu_r' - i\mu_r'')$. In this description, details of the field behavior on the scale of scatterer size and separation are lost, and often are not of practical interest.

For passive materials, $\mu_r'' \geq 0$ and $\epsilon_r'' \geq 0$. As for the real parts of the material parameters, for many common materials μ_r' and ϵ_r' are positive, but there are exceptions. For example, in plasmas the combination of ordinary displacement current density with electron-convection current density can yield a net negative real part of the permittivity for sufficiently low frequencies [1, pp. 309–319]. Indeed, Rotman [11] has shown how an artificial dielectric can reproduce such a negative permittivity and serve as an equivalent model for a plasma. A transmission line equivalent circuit for describing the negative ϵ_r' of a plasma is discussed in [12]. Negative permittivity also appears near a resonance frequency in Lorentz's theory of dispersion (see [13], for example).

When one (but not both) of ϵ_r' or μ_r' is negative, plane waves decay exponentially, like modes below cutoff in a waveguide. However, when both ϵ_r' and μ_r' are negative, waves can still propagate in such a medium since the product $\mu\epsilon$ remains positive. In this case, we have a "backward wave," for which the phase of the wave moves in the direction opposite from that of the energy flow. For lossless media, this means that the phase velocity and group velocity have opposite signs. Many authors have attributed the first study of such media to Veselago [14] in 1967, but Sivukhin [15] in 1957 examined briefly their properties; both of them as well as Malyuzhinets [16] and Silin [17]–[19] give credit to much earlier work of Mandel'shtam [20], [21]. Mandel'shtam himself referred to a 1904 paper of Lamb [22], who may have been the first person to suggest the existence of backward waves (his examples involved mechanical systems rather than electromagnetic waves). In his 1904 book on optics, Schuster [23] briefly notes Lamb's work, and gives a speculative discussion of its implications for optical refraction, should a material with such properties ever be found. In 1905, Pocklington [24] anticipated the work of Malyuzhinets by almost 50 years, showing that in a specific backward-wave medium, a suddenly activated source produces a wave whose

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[1]Scatterers of complex geometry which can be resonant may result in an anisotropic medium, for which ϵ and μ are tensors, or even in a *bianisotropic* medium, for which $\mathbf{D}$ and $\mathbf{H}$ are each affected by both $\mathbf{E}$ and $\mathbf{B}$. We limit our attention in this paper to isotropic, nonbianisotropic materials.

group velocity is directed away from the source, while its phase velocity moves toward the source. More recently, many other authors [25]–[32] have studied the properties and potential applications of negative-index materials in detail.

A medium composed of periodically placed scatterers generates polarization and magnetization densities. The densities are related to the distribution of the scatterers and their polarizabilities. As a result, a wave propagating through an array of these scatterers will see the material as an effective medium. The problem of effective-medium theory and modeling the electromagnetic response of inclusions embedded in a host material has a long history going back to Maxwell and Rayleigh [3]–[10]. One notable work is that of Lewin [7], who incorporates the solution of a boundary-value problem for scattering by a sphere in a unit cell and then assumes the medium is composed of a large number of these cells and thereby obtains a description in terms of effective-medium parameters $\mu_{\rm e}$ and $\epsilon_{\rm e}$. When the size of the spherical scatterers is not small compared to a wavelength *in the material of the scatterers* (but *is* small compared to a wavelength in the matrix material), $\mu_{\rm e}$ and $\epsilon_{\rm e}$ are frequency-dependent. Recently, several papers have studied the problem of designing engineered artificial materials with negative μ_r' and ϵ_r' formed from periodic arrays of unusually-shaped conducting scatterers [33]–[44]. In practice, these structures are quite complicated to fabricate.

In this paper, we use the work of Lewin to illustrate that, over specific frequency bands, both the effective ϵ_r' and μ_r' of a material composed of insulating spherical particles embedded in a matrix can become simultaneously negative. We will show that the properties of an array of spherical particles can behave in a way similar to that of an array of geometrically more complicated conducting scatterers, and that the effective electric and/or magnetic polarizabilities of both types of scatterer have the same characteristic of exhibiting a resonance, hence resulting in an effective negative ϵ_r' and μ_r' in certain frequency ranges. This result suggests the possibility of developing double negative (DNG) (or negative-index) materials that could be fabricated much more simply than those that have been proposed up until now.

II. Effective Dielectric and Magnetic Properties of a Spherical-Particle Composite

In 1947, Lewin [7] derived an expression for the effective properties of an array of spherical particles embedded in a background matrix, see Fig. 1. Ten years later Khizhniak wrote a series of papers [8]–[10] in which he generalized Lewin's model and presented expressions for the effective material-property tensors of an artificial material formed by an array of scatterers with arbitrary geometric shapes. For the array of lossless magnetodielectric spheres, the relative effective $\mu_{\rm re}'$ and $\epsilon_{\rm re}'$ for the geometry shown in Fig. 1 are given by [7]

$$\epsilon_{\rm re}' = \epsilon_{r1}\left(1+\frac{3v_f}{\frac{F(\theta)+2b_{\rm e}}{F(\theta)-b_{\rm e}}-v_f}\right) \tag{1}$$

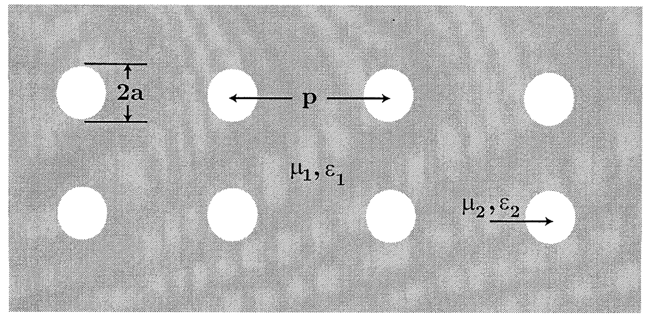

Fig. 1. Composite structure containing spherical particle.

and

$$\mu_{\rm re}' = \mu_{r1}\left(1+\frac{3v_f}{\frac{F(\theta)+2b_m}{F(\theta)-b_m}-v_f}\right). \tag{2}$$

In these expressions, μ_{r1} and ϵ_{r1} are the relative permeability and permittivity of the matrix (host) medium, μ_{r2} and ϵ_{r2} are the relative permeability and permittivity of the inclusions, and

$$b_{\rm e} = \frac{\epsilon_1}{\epsilon_2}, \qquad b_m = \frac{\mu_1}{\mu_2}. \tag{3}$$

The volume fraction v_f of the spherical inclusions is given by

$$v_f = \frac{4\pi a^3}{3p^3} \tag{4}$$

where a is the particle radius and p is the particle spacing. The function $F(\theta)$ is

$$F(\theta) = \frac{2(\sin\theta - \theta\cos\theta)}{(\theta^2-1)\sin\theta + \theta\cos\theta} \tag{5}$$

where

$$\theta = k_0 a\sqrt{\epsilon_{r2}'\mu_{r2}'} \tag{6}$$

and the free-space wavenumber is $k_0 = 2\pi/\lambda$, where λ is the free-space wavelength. These formulas were rediscovered later by Granqvist [45, eq. (3), (9) and (12)], Waterman and Pedersen [46, eq. (33)] (if the small imaginary term of order $(k_1 a)^3$ in our notation is removed from (21b) and (21c) for $n = 1$), by Mahan [47, eq. (5)] and by Sarychev *et al.* [48, eqs. (12), (15), and (21)].

The dependence of $F(\theta)$ versus θ for real θ is shown in Fig. 2. Notice the resonant nature of this function, which becomes infinite at certain frequencies, and is negative in some (relatively small) ranges of θ. We then ask: can this result in DNG behavior? This question can be answered by investigating (1) and (2) as functions of θ. From (1), we see that $\epsilon_{\rm re}' < 0$ if

$$\frac{3v_f}{\frac{F(\theta)+2b_{\rm e}}{F(\theta)-b_{\rm e}}-v_f} < -1. \tag{7}$$

If we assume that $b_{\rm e} > 0$ (as it will be if both matrix and inclusion are made from ordinary dielectrics), then this can be rearranged to give the following condition on $F(\theta)$ to guarantee that $\epsilon_{\rm re}' < 0$

$$-b_{\rm e}\frac{2+v_f}{1-v_f} < F(\theta) < -2b_{\rm e}\frac{1-v_f}{1+2v_f}. \tag{8}$$

Since $0 \le v_f < \pi/6$ (the maximum occurs when the spheres touch), it is necessary (but not sufficient) that $F(\theta)$ be negative

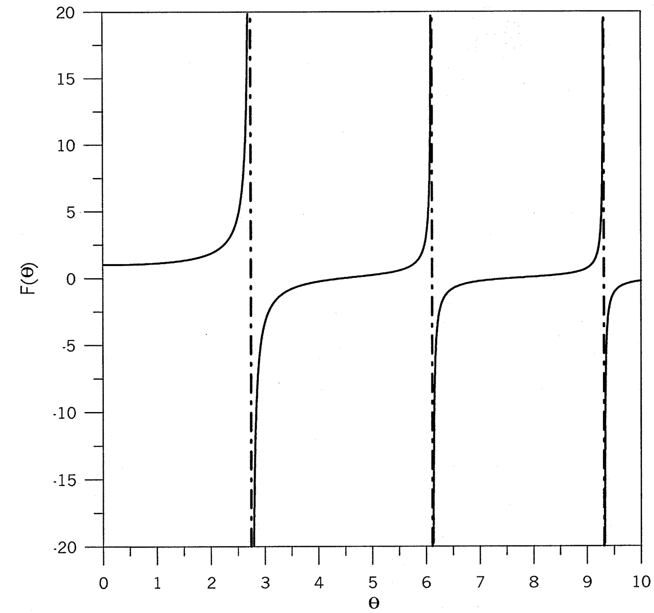

Fig. 2. Functional behavior of $F(\theta)$ versus θ. The dashed-dot lines represent the asymptotes.

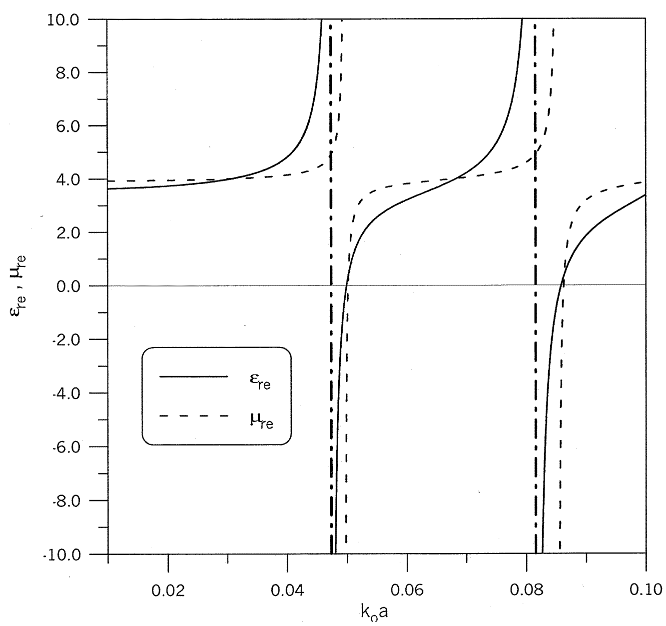

Fig. 3. $\epsilon_{\rm re}$ and $\mu_{\rm re}$ for $v_f = 0.5, \epsilon'_{r1} = \mu'_{r1} = 1, \epsilon'_{r2} = 40$, and $\mu'_{r2} = 200$. The dashed-dot lines represent the asymptotes.

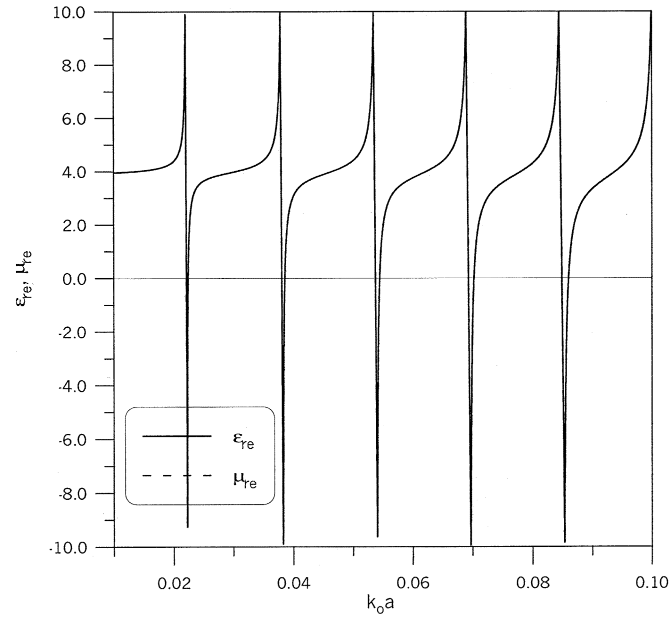

Fig. 4. $\epsilon_{\rm re}$ and $\mu_{\rm re}$ for $v_f = 0.5$, $\epsilon'_{r1} = \mu'_{r1} = 1$, $\epsilon'_{r2} = 200$, and $\mu'_{r2} = 200$. Notice that $\epsilon_{\rm re}$ and $\mu_{\rm re}$ are identical.

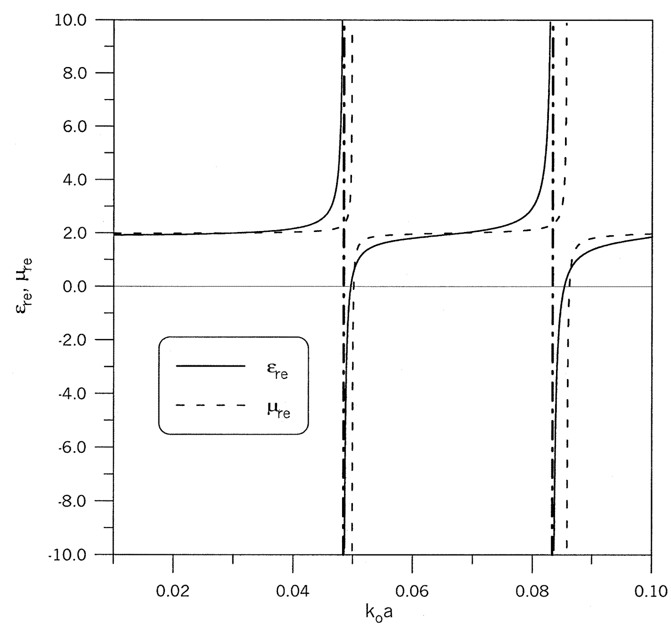

Fig. 5. $\epsilon_{\rm re}$ and $\mu_{\rm re}$ for $v_f = 0.25$, $\epsilon'_{r1} = \mu'_{r1} = 1$, $\epsilon'_{r2} = 40$, and $\mu'_{r2} = 200$. The dashed-dot lines represent the asymptotes.

to have $\epsilon'_{\rm re} < 0$. From Fig. 2, we see that this is possible. Similarly, starting with (2), we find that the condition in (8) (now with b_m in place of $b_{\rm e}$) will guarantee that $\mu'_{\rm re} < 0$. The possibility of having negative $\epsilon'_{\rm re}$ and $\mu'_{\rm re}$ in this type of composite structure is implicit in Lewin's work [7], was specifically alluded to by Khizhniak [9], and was demonstrated explicitly in [46]–[49].

III. Numerical Examples

In this section, we present several examples that demonstrate the feasibility of achieving negative $\mu_{\rm re}$ and $\epsilon_{\rm re}$ for such a composite material, and show how the frequency range for this behavior depends on ϵ'_{r1}, ϵ'_{r2}, μ'_{r1}, μ'_{r2}, and v_f. Fig. 3 shows results for $v_f = 0.5$, $\epsilon'_{r1} = \mu'_{r1} = 1$, $\epsilon'_{r2} = 40$, and $\mu'_{r2} = 200$ as a function of $k_0\, a$. Between $0 \le k_0\, a \le 0.1$ there are two regions where both $\mu'_{\rm re}$ and $\epsilon'_{\rm re}$ become negative. Over a portion of each of these two regions, $\mu'_{\rm re}$ and $\epsilon'_{\rm re}$ become negative simultaneously, producing a negative-index material. It is possible to have $\epsilon'_{\rm re}$ negative over the same region where $\mu'_{\rm re}$ is negative by having $\epsilon'_{r1}/\epsilon'_{r2} = \mu'_{r1}/\mu'_{r2}$. This is illustrated in Fig. 4 for $v_f = 0.5$, $\epsilon'_{r1} = \mu'_{r1} = 1$, $\epsilon'_{r2} = 200$, and $\mu'_{r2} = 200$.

Decreasing the inclusion volume fraction v_f has the effect of narrowing the band of frequencies for which $\epsilon'_{\rm re}$ and $\mu'_{\rm re}$ become negative. This is seen by comparing the results in Figs. 3 and 5. The results in Fig. 5 are for the same materials as in Fig. 3, but v_f is reduced to 0.25 from 0.5. The bandwidth for which a negative $\epsilon'_{\rm re}$ is obtained is 5.5% in Fig. 3 and 2.5% in Fig. 5.

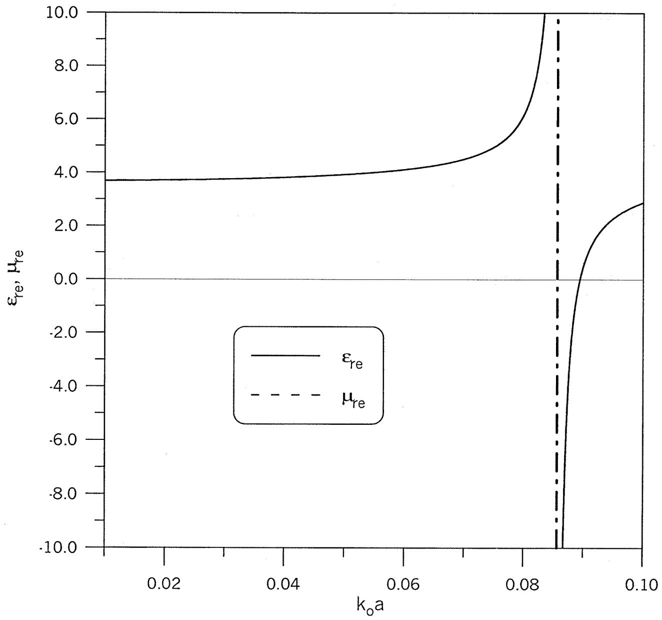

Fig. 6. $\epsilon_{\rm re}$ and $\mu_{\rm re}$ for $v_f = 0.5$, $\epsilon'_{r1} = \mu'_{r1} = 1$, $\epsilon'_{r2} = 50$, and $\mu'_{r2} = 50$. The dashed-dot line represents the asymptote.

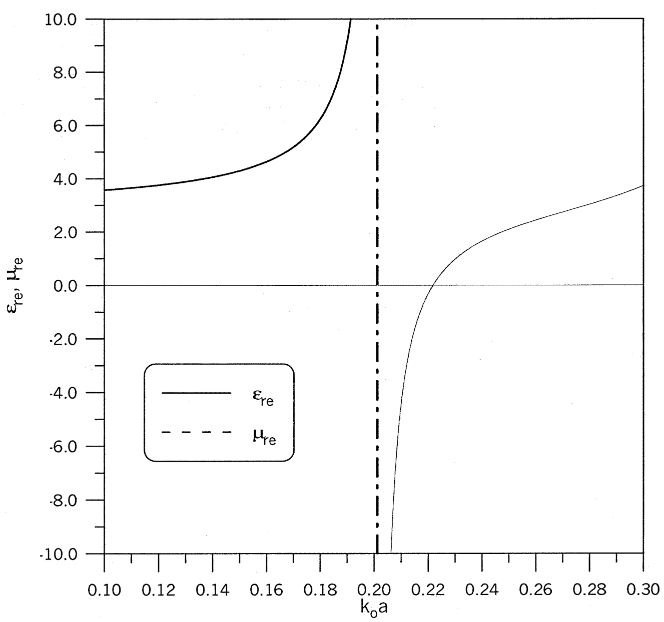

Fig. 7. $\epsilon_{\rm re}$ and $\mu_{\rm re}$ for $v_f = 0.5$, $\epsilon'_{r1} = \mu'_{r1} = 1$, $\epsilon'_{r2} = 20$, and $\mu'_{r2} = 20$. The dashed-dot line represents the asymptote. Notice that $\epsilon_{\rm re}$ and $\mu_{\rm re}$ are identical.

Besides v_f, the product of ϵ'_{r2} and μ'_{r2} influences the bandwidth and location of the resonance. This is seen by comparing Figs. 3 and 4. The bandwidth in Fig. 3 is 5.5% and is broader than in Fig. 4, which is 1.1%. In Fig. 3, $\epsilon'_{r2}\,\mu'_{r2} = 8000$, and in Fig. 4, $\epsilon'_{r2}\,\mu'_{r2} = 40\,000$. Also notice that the first resonance in Fig. 3 occurs at $k_0 a = 0.048$, while the first resonance in Fig. 4 occurs at $k_0 a = 0.022$. Thus, by making the product of ϵ'_{r2} and μ'_{r2} smaller, the first resonance is moved to larger values of $k_0\,a$ and the frequency bandwidth over which the permittivity and permeability are negative increases. We see this by comparing Figs. 3, 6 and 7. The results in Fig. 6 correspond to $\epsilon'_{r2}\,\mu'_{r2} = 2500$; the results in Fig. 7 correspond to $\epsilon'_{r2}\,\mu'_{r2} = 400$. The location of the first resonance and its bandwidth are inversely proportional to the product of ϵ'_{r2} and μ'_{r2}. The bandwidth for the example shown in Fig. 7 is 10%.

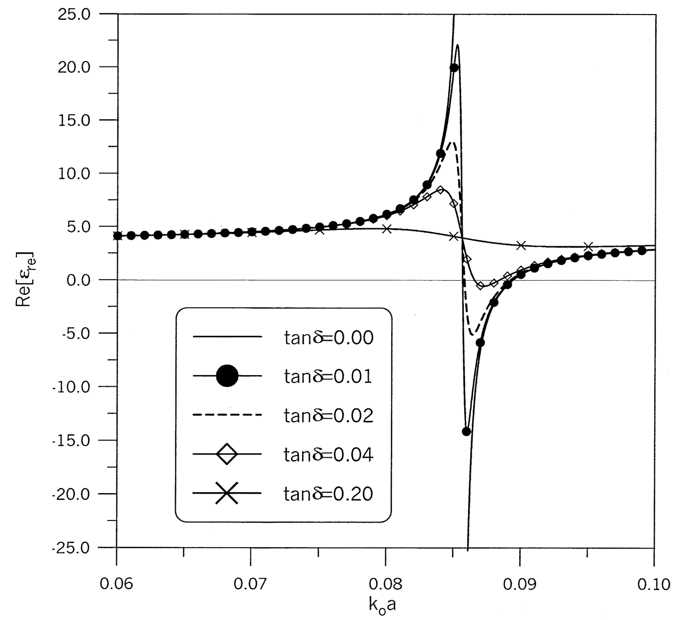

Fig. 8. $\epsilon'_{\rm re}$ for $v_f = 0.5$, $\epsilon'_{r1} = \mu'_{r1} = 1$, $\epsilon'_{r2} = 50$, and $\mu'_{r2} = 50$.

A. Lossy Materials

In Lewin's original work [7], the permeability and permittivity of the inclusions were allowed to be complex, i.e., to exhibit loss. In fact, all realistic materials have some loss. Losses have the effect of damping out the resonant behavior of the composite. Since for large values of particle loss (as considered by Lewin [7] and others [50], [51]), no such resonance is exhibited, we anticipate that for some threshold value of the loss, $\mu'_{\rm re}$ and $\epsilon'_{\rm re}$ will always remain positive. Unfortunately, it does not seem possible to analytically determine what this threshold value of loss will be for any given values of ϵ'_{r1}, ϵ'_{r2}, μ'_{r1}, μ'_{r2} and v_f. We can, however, illustrate the effects of losses for specific examples by allowing ϵ_2 and/or μ_2 to become complex.

We show this effect in Fig. 8, for $\epsilon'_{r1} = 1$, $\epsilon'_{r2} = 50$, $\mu'_{r1} = 1$, $\mu'_{r2} = 50$ and $v_f = 0.5$. In this figure, the dependence of the real part of the effective permittivity on normalized frequency is shown for several different values of the dielectric loss tangent of the inclusions, defined as $\tan\delta = \epsilon''_{r2}/\epsilon'_{r2}$. The dielectric loss tangent of the matrix, as well as the magnetic loss tangents of both materials, is taken to be zero: $\epsilon''_{r1} = \mu''_{r1} = \mu''_{r2} = 0$. Notice that for this example, the real part of the effective permittivity can still be negative for loss tangents as large as 0.04. However, for larger values of $\tan\delta$ the resonance is damped out and the real part of the effective permittivity remains positive. This shows that if the inclusion (i.e., the spherical particle) becomes too lossy, DNG properties cannot be realized.

IV. Coated Particles

In order to achieve some desired effective electromagnetic properties of a composite medium, the simple array of homogeneous spherical particles may be impractical, because no material exists which simultaneously exhibits the required values of ϵ'_{r2} and μ'_{r2}. It should be possible to circumvent this problem by making the particle itself out of a composite material. In [52], for example, the work of Lewin was extended to handle the case when the spherical inclusions are spheres of one material coated with a layer of different material. Formulas for the effective electromagnetic properties of arrays of such coated particles are given in [52]. In fact, this coated dielectric-metallic particle concept was the basis of the work presented in [37]. However, the work of [37] is limited to a quasistatic approximation, while the work in [7] and [52] accounts properly for the resonant behavior of the fields inside the spherical particles.

V. Other Types of Arrays of Resonant Inclusions

From the viewpoint of scattering theory, all scattering objects can be represented by effective electric and/or magnetic polarizability densities. If these polarizability densities exhibit a characteristic resonant behavior with frequency, then it should be possible to obtain negative effective permeability and permittivity from composite structures more general than cubical arrays of spherical particles. Khizhniak's generalization [8]–[10] of Lewin's work does indeed suggest this.

Two-dimensional (2-D) arrays of magnetodielectric cylinders should also be capable of exhibiting DNG behavior (albeit in an anisotropic fashion). Indeed, in 1959 Khizhniak [53] extended his previous work to present expressions for the effective permeability and permittivity of arrays of magnetodielectric rods, infinitely extended along their axes. This result has also been rediscovered over the years, for example in [54]. It should come as no surprise that the expressions given in these papers have a functional form similar to those of Lewin [7] and Khizhniak [8]–[10] for three-dimensional (3-D) arrays. The expressions for the rods exhibit a characteristic resonant behavior and result in negative effective permeability and permittivity.

A recent paper by O'Brien and Pendry [55] analyzes the same rod-based composite structure as Khizhniak [53] and Matagne [54], authors in [55] were apparently unaware of these earlier works. In [55], the effective permeability is expressed in terms of integrals involving Bessel functions, using an argument based on the field scattered by a single rod. The integrals in their expression can actually be evaluated in closed form, giving a formula for the effective permeability similar to, but not exactly the same as, that obtained by Khizhniak [53]. The two different expressions approach one another only in the limit of very small volume fractions. We believe that the method of [55] is in error, which can be traced to the fact that the field acting on an individual rod embedded in an array was not properly modeled. Numerical comparisons of the two results show large differences between predicted values of effective permittivity and permeability, calling the validity of the formulas of [55] into question.

VI. Comments of the Validity of Expressions for the Effective Properties

In the derivation of his expressions for the effective material properties of the artificial dielectric, Lewin [7] offered some empirical guidelines for determining whether or not the formulas would be accurate for given values of the structural parameters.

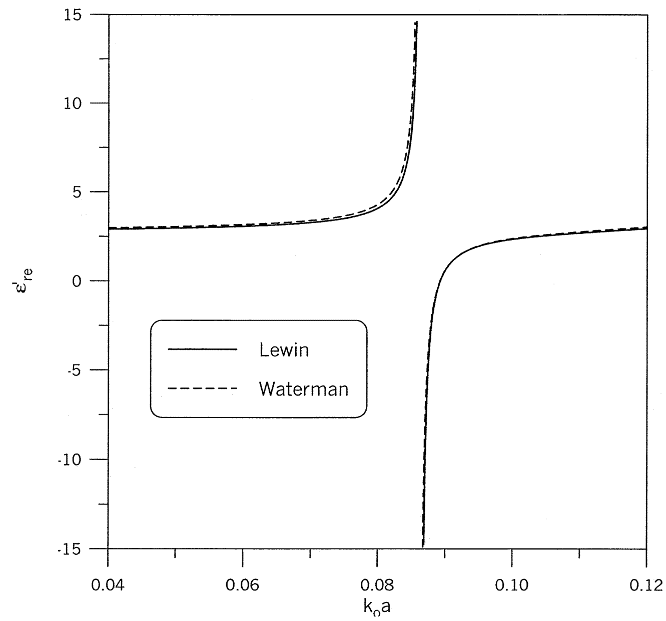

Fig. 9. $\epsilon'_{\rm re}$ for $v_f = 0.4$, $\epsilon'_{r1} = \mu'_{r1} = 1$, $\epsilon'_{r2} = 50$, and $\mu'_{r2} = 50$.

It seems to us that his conditions are perhaps overly conservative, and not entirely based upon physical considerations of the structure.

One of the conditions for validity of (1) and (2) should be the sufficiency of using only the dipole terms outside the inclusions in representing the total field there. This in turn would place a restriction that the volume fraction v_f of the inclusions should not be too large. Waterman and Pedersen [46] have obtained Lewin's results as a special case of their more general analysis, which could in principle retain as many higher-order multipole terms in the field representation as desired. In Figs. 9–11 we illustrate the effects of including higher order (up to seventh) multipole effects based on [46, eqs. (35a)-(35b)] (again, with the small imaginary terms in their eqs. (21b) and (21c) omitted, and with the misprinted term $(3f/4\pi\alpha)^{7/8}$ corrected to read $(3f/4\pi\alpha)^{7/3}$ [56]). For a small volume fraction where $v_f = 0.1$, a comparison of Waterman's and Lewin's results shows no graphically discernable difference. For a larger volume fraction $v_f = 0.4$, we see from Fig. 9 that a small difference appears near resonance, but that overall Lewin's approximation is still quite accurate. When $v_f = 0.5$ (the spherical inclusions are almost touching), Fig. 10 shows that the multipole terms serve to increase the predicted effective permittivity somewhat by comparison with the Lewin formula. Despite this, the resonance still occurs at virtually the same frequency, and the bandwidth over which $\epsilon_{\rm re}$ is negative is actually increased somewhat. Fig. 11 shows the dependence of the two formulas on filling fraction when frequency is held constant (and away from resonance). We see that the results that include higher multipoles differ noticeably from the Lewin prediction only for $v_f > 0.4$. Even at $v_f = 0.5$, the error in this example is less than 10%. Based on these comparisons, we can conclude that (1) and (2) are certainly accurate when $v_f < 0.4$. Even as the volume fraction approaches 0.5, Lewin's formula is still accurate enough for many purposes. If additional precision is needed, Waterman's formula can always be used.

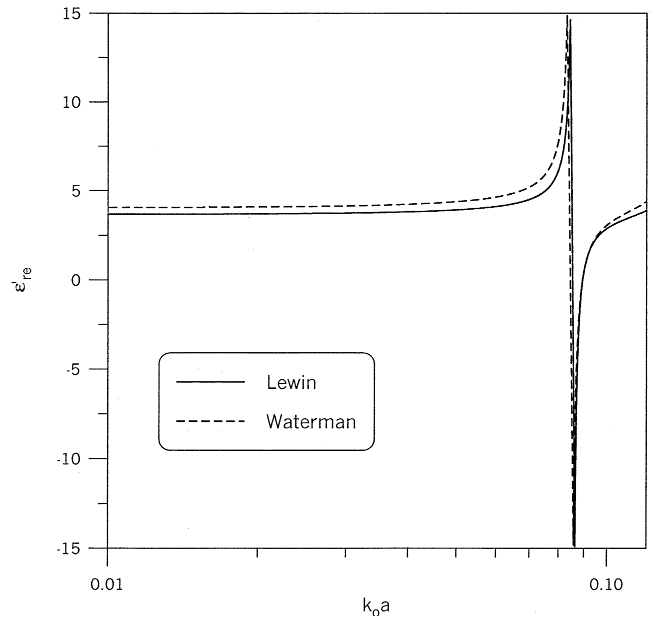

Fig. 10. $\epsilon'_{\rm re}$ for $v_f = 0.5$, $\epsilon'_{r1} = \mu'_{r1} = 1$, $\epsilon'_{r2} = 50$, and $\mu'_{r2} = 50$.

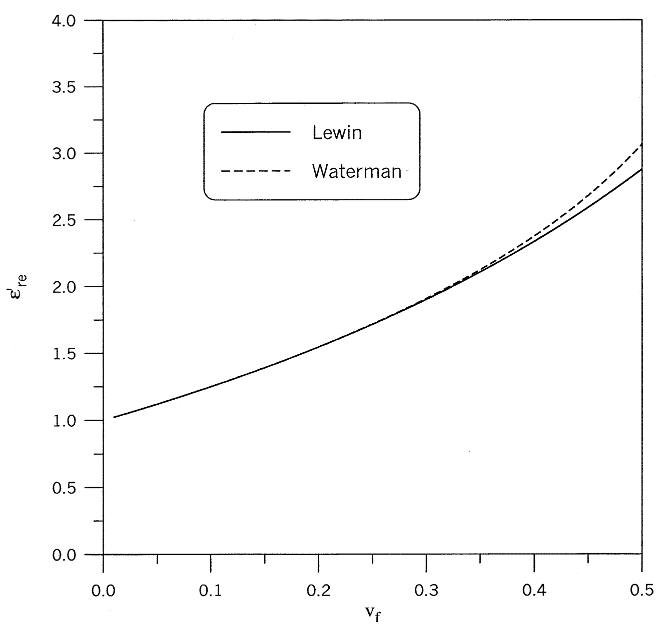

Fig. 11. $\epsilon'_{\rm re}$ for $k_0 a = 0.1$, $\epsilon'_{r1} = \mu'_{r1} = 1$, $\epsilon'_{r2} = 50$, and $\mu'_{r2} = 50$.

The other condition for Lewin's formulas to be accurate is somewhat more fundamental. The very notion of an effective medium describable by the parameters $\epsilon_{\rm e}$ and $\mu_{\rm e}$ requires that only one Floquet-Bloch mode of this periodic structure is capable of propagation; that is, we require that $k_1 p < \pi$. It is possible that an even stronger condition may be necessary here, as the derivation of the expressions for the effective material properties implies that the variation of the average field over a spatial period should be small. In other words, we should have the (possibly) stronger condition $k_{\rm e} p < 1$, say, where $k_{\rm e} = \omega\sqrt{\mu_{\rm e}\epsilon_{\rm e}}$ is the wavenumber in the effective medium. This can be compared with Lewin's condition, which we may write as $k_{\rm e} p < 1/v_f$. This condition seems too restrictive, especially for small volume fractions. It is difficult to make this condition more precise or rigorous at this point. Variations of the multiple-scale homogenization procedure such as that used for the heat equation in [57] and [58] may prove fruitful in this regard.

It should also be noted that the importance of the effective medium parameters is not only to predict the correct propagation constant in the composite material, but the correct wave impedance as well, so that reflection and transmission coefficients are accurately determined. When the inclusions are dense electrically and/or magnetically as is the case here, it is crucial to define the average fields in such a way that this occurs. The results of [59] strongly suggest that the Lewin definitions of $\mu_{\rm e}$ and $\epsilon_{\rm e}$ are correct in this sense too, lending further support to their validity.

VII. Conclusion

In this paper, we show how a composite medium realized by an array of spherical particles embedded in a background matrix can yield an effective negative permeability and permittivity. From a scattering-theory viewpoint, negative effective permeability and permittivity of a composite structure are possible if the effective electric and/or magnetic polarizabilities exhibit a characteristic resonant behavior, hence, it should be no surprise that the spherical particles, and any magnetodielectric inclusion for that matter, behave in the same manner as arrays of more complicated conducting scatterers. The type of composite material discussed in this paper introduces a new class of potential DNG materials. In this approach no complicated metallic scatterers are required and the composite based on a spherical-particle array has the added advantage of being isotropic. This approach can be readily extended to other geometries and to other types of inclusions. Work is currently underway to fabricate and experimentally verify samples of this new type of DNG medium.

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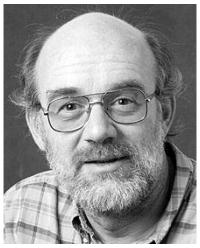

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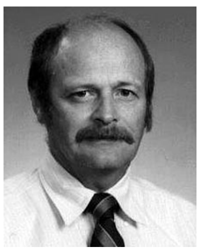

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