Restoring the physical meaning of metamaterial constitutive parameters

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Metamaterial homogenization is often based on implicit assumptions inspired by natural material models. However, retrieved permittivity and permeability frequently retain nonphysical values, especially near the metamaterial resonances where most interesting features are expected. We explain here the nature of typical homogenization artifacts, relating them to an inherent form of magnetoelectric coupling associated with the finite phase velocity along metamaterial arrays. Our findings allow restoring the proper definition and physical meaning of local constitutive parameters for metamaterials.

Negative-index metamaterials (NIMs), ε-near-zero and ε-very-large metamaterials,[1] i.e., artificial materials supporting negative refraction or anomalous values of permittivity, have been theoretically shown to possess features not available in nature and ideal for radiation, imaging, cloaking, and waveguiding applications. From the theoretical standpoint, such anomalous properties may revolutionize several applied fields, but the same definition of metamaterial constitutive parameters has proven to be challenging, in particular when extreme (very large, very low, or negative) values are considered.[2,3] Several homogenization models have been put forward to macroscopically describe the wave propagation in metamaterials, with the goal of treating complex arrays of resonant inclusions as bulk materials.[3–11] The most common definition of homogenized metamaterial parameters is based on retrieval methods,[12] which implicitly postulate that a metamaterial may be described as a natural material, with local effective permittivity and permeability, and aims at extracting these parameters from scattering measurements. This method cannot ensure that the extracted parameters have a proper physical meaning, as it implicitly relies on the validity of the chosen model. Indeed, the metamaterial parameters retrieved from experiments within such schemes often do not satisfy basic passivity and causality constraints[12,13] required by the second law of thermodynamics and by causality, energy conservation, and the Kramers-Kronig relations[14]

$$\mathrm{Im}[\varepsilon_{\mathrm{eff}}] > 0, \quad \partial\varepsilon_{\mathrm{eff}}/\partial\omega \geqslant 0, \quad \partial(\omega\varepsilon_{\mathrm{eff}})/\partial\omega \geqslant 0, \quad (1)$$

and similarly for the effective permeability μ_{eff}, under an $e^{-i\omega t}$ time convention.

In particular, frequency bands in which one of the two retrieved parameters experiences an unphysical "antiresonant response" with a negative slope and a negative imaginary part[12] are common in metamaterial retrieval procedures. These artifacts have often been justified with creative but unpersuasive arguments, generically associated with spatial dispersion, influence of higher-order multipoles, neglected bianisotropic effects in the inclusions, and other related issues.[12] The presence of these antiresonances has been verified in dozens of papers on metamaterial characterization and experiments, and it has been accompanied by serious doubts on whether the same meaning of such extreme metamaterial parameters, which go beyond what is commonly available in nature, is acceptable. In particular, the same possibility to define

metamaterial parameters is doubted near the array resonances, where the most interesting effects usually take place. In the following, we thoroughly address this issue, showing that these antiresonance artifacts may be traced back to a weak form of spatial dispersion effects associated with the finite phase velocity along the metamaterial array, which is usually neglected in metamaterial homogenization. Properly taking into account these effects, we put forward a way to restore physically meaningful local metamaterial parameters that may properly describe the exotic properties of metamaterials even in regimes associated with extreme or anomalous parameters.

In Ref. 11, we have proposed a general analytical homogenization method that can rigorously describe the wave interaction with periodic metamaterial arrays formed by arbitrary magnetodielectric inclusions. For simplicity, let us consider here the special situation in which the array is formed by a cubic lattice with period d, much smaller than the wavelength of operation λ_0, made of center-symmetric inclusions with no bianisotropic effects. In such case, the inclusions may be effectively described by their electric and magnetic scalar polarizabilities α_e and α_m, which relate the electric and magnetic dipole moments to the local electric and magnetic fields at their center. This is the most ideal situation to homogenize metamaterial arrays, and it is widely believed that a simple isotropic model based on scalar permittivity and permeability should be accurate to describe a metamaterial under these conditions. A rigorous analysis of the coupling among the inclusions,[11] however, shows that the effective constitutive relations should be written, for an arbitrary $e^{i\boldsymbol{\beta}\cdot\mathbf{r}}$ space variation, where $\boldsymbol{\beta}$ is the Bloch-wave vector, as

$$\begin{aligned}
\mathbf{D}_{\mathrm{av}} &= \varepsilon_0\mathbf{E}_{\mathrm{av}} + \mathbf{P}_{\mathrm{av}} = \varepsilon_{\mathrm{eff}}\mathbf{E}_{\mathrm{av}} - \kappa_{\mathrm{eff}}\boldsymbol{\beta}\times\mathbf{H}_{\mathrm{av}}, \\
\mathbf{B}_{\mathrm{av}} &= \mu_0\mathbf{H}_{\mathrm{av}} + \mathbf{M}_{\mathrm{av}} = \mu_{\mathrm{eff}}\mathbf{H}_{\mathrm{av}} + \kappa_{\mathrm{eff}}\boldsymbol{\beta}\times\mathbf{E}_{\mathrm{av}},
\end{aligned} \quad (2)$$

where closed-form analytical expressions for the effective constitutive parameters $\varepsilon_{\mathrm{eff}}$, μ_{eff}, and κ_{eff} have been derived in Refs. 11 and 15 and, in the present scenario, the average fields are defined as $\mathbf{F}_{\mathrm{av}} = \frac{1}{d^3}\int_V \mathbf{F}(\mathbf{r})e^{-i\boldsymbol{\beta}\cdot\mathbf{r}}\,d\mathbf{r}$, with V being the unit-cell volume, in analogy with Ref. 10. Due to the array and inclusion symmetries, the constitutive parameters are indeed isotropic, but an inherent form of magnetoelectric coupling at the lattice level, represented by κ_{eff}, arises despite the assumed symmetries. This coupling, which is consistent with recent homogenization studies,[8,9] is related to the asymmetry introduced by the finite phase velocity along the array

$v_p = \omega/\beta$ (Ref. 11) and represents a weak form of spatial dispersion. These expressions may be generalized with proper tensorial notation to anisotropic inclusions and lattices, and to arbitrary propagation direction, still retaining this relevant form of weak spatial dispersion; this goes, however, beyond the scope of the present paper in which, for clarity, we limit our analysis to the isotropic scenario. We will prove in the following that the antiresonant and nonphysical artifacts associated with retrieved parameters near the inclusion resonances may be directly related to these second-order spatial-dispersion effects, inherent to lattice propagation, and relevant even in the long-wavelength regime $d \ll \lambda_0$.

The improved constitutive model (2) has been proven in Ref. 11 to be valid for any pair $(\boldsymbol{\beta}, \omega)$, totally independent of the local-field distribution along the array, of the possible presence of external sources, and of the local impedance ratio $E_{\mathrm{av}}/H_{\mathrm{av}}$, ensuring that three local parameters $\varepsilon_{\mathrm{eff}}$, μ_{eff}, and κ_{eff} are sufficient to properly describe the metamaterial properties in the long-wavelength limit for any form of excitation. In retrieval experiments, however, we usually extract the eigenmodal response of the array, i.e., we operate in absence of impressed sources inside the metamaterial. In such a case, the average fields satisfy, using Eq. (2),

$$i\boldsymbol{\beta} \times \mathbf{E}_{\mathrm{av}} = i\omega \frac{\mu_{\mathrm{eff}}}{1 - \omega\kappa_{\mathrm{eff}}} \mathbf{H}_{\mathrm{av}} = i\omega\mu_{\mathrm{eq}}\mathbf{H}_{\mathrm{av}},$$
$$i\boldsymbol{\beta} \times \mathbf{H}_{\mathrm{av}} = -i\omega \frac{\varepsilon_{\mathrm{eff}}}{1 - \omega\kappa_{\mathrm{eff}}} \mathbf{E}_{\mathrm{av}} = -i\omega\varepsilon_{\mathrm{eq}}\mathbf{E}_{\mathrm{av}}, \quad (3)$$

which allows defining *equivalent* constitutive parameters $\varepsilon_{\mathrm{eq}}$ and μ_{eq} (Ref. 11), related to the effective permittivity and permeability through the normalization factor $(1 - \omega\kappa_{\mathrm{eff}})$. The eigenmodal assumption forces a specific ratio $E_{\mathrm{av}}/H_{\mathrm{av}}$ in the metamaterial, i.e., the modal characteristic impedance, which lets us write the constitutive relations as in a local isotropic material, analogous to the assumption commonly made in retrieval experiments. However, these *equivalent* parameters inherently contain a hidden form of weak spatial dispersion associated with a finite κ_{eff}, and for this reason they cannot be considered local even in the long-wavelength limit for which the *effective* parameters are. They are expected to inherently depend on the excitation, on the local ratio $E_{\mathrm{av}}/H_{\mathrm{av}}$, and on the direction of propagation. It is not surprising, therefore, that their dispersion may not satisfy basic physical constraints, as in Eq. (1), and their same physical meaning, as an averaged electric or magnetic polarizability of the array, is compromised for $\beta, \omega \neq 0$.

In order to see how these effects play an important role in metamaterial homogenization, consider first the simple metamaterial geometry formed by dielectric spheres with permittivity $\varepsilon = 120\varepsilon_0$, permeability $\mu = \mu_0$, and normalized radius $a/d = 0.45$ (an analogous array was considered in Ref. 11 to analyze the spatial dispersion effects in the homogenization of resonant metamaterials). The idea of using a dense array of high-index spheres to produce isotropic negative effective parameters has been put forward in Refs. 6 and 16, and it indeed provides an interesting venue to verify the effect of κ_{eff}, even in cases for which ideal symmetries and isotropy would suggest the absence of significant magnetoelectric coupling effects in metamaterial homogenization.

Figure 1 shows the normalized α_e and α_m (dark solid and dashed lines, respectively), as a function of $k_0 d$, with
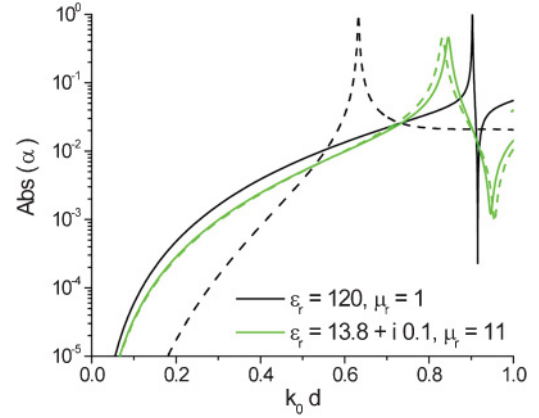

FIG. 1. (Color online) Magnitude of the electric (solid) and magnetic (dashed) polarizability coefficients for spheres with: $\varepsilon = 120\varepsilon_0$, $\mu = \mu_0$, $a/d = 0.45$ (darker, black); $\varepsilon = (13.8 + i0.1)\varepsilon_0$, $\mu = 11\mu_0$, $a/d = 0.4$ (lighter, green).

$k_0 = 2\pi/\lambda_0$. Their value has been normalized to $k_0^3/(6\pi)$, ensuring that their peak magnitude (at resonance) is unity in absence of Ohmic absorption. As expected, the magnetic response is much weaker than the electric one for longer wavelengths, but the first dipolar resonance is magnetic in nature, arising at $k_0 d = 0.63$, followed by an electric one at $k_0 d = 0.9$. The corresponding k_0-β dispersion diagram is reported in Ref. 17, highlighting the presence of a large band gap around the magnetic resonance (shadowed region).

Figure 2 shows the calculated effective and equivalent parameters for such an array as a function of frequency for eigenmodal propagation, using the rigorous analytical formulation (2) derived in Ref. 11. The shadowed region highlights also here the magnetic band-gap region. In the low-frequency regime, well below the first band gap and for very small values of βd and $k_0 d$, equivalent and effective parameters coincide; they are both positive and practically constant with frequency. In this limit, classic homogenization techniques apply very well, and the metamaterial behaves as a regular mixture. Closer to the band gap, effective and equivalent permeability both drastically increase, supporting a typical Lorentzian dispersion associated with the magnetic resonance of the inclusions. However, near this resonance the *equivalent* permittivity experiences an anomalous antiresonant dispersion, a typical feature of the permittivity retrieved near a magnetic resonance with simple homogenization schemes.[12] The *effective* permittivity, on the contrary, has positive slope and a physical Lorentzian response. It is evident that the *effective* permittivity describes a physical quantity, relating the local $\mathbf{E}_{\mathrm{av}}$ to $\mathbf{P}_{\mathrm{av}}$ as in Eq. (2), without mixing in the effect of $\mathbf{H}_{\mathrm{av}}$ associated with the magnetoelectric coupling, contrary to the corresponding *equivalent* parameter.

It is interesting to analyze the reasons behind the large divergence between effective and equivalent permittivity in this regime: at the lower edge of the band gap $k_0 d = 0.59$, near the magnetic resonance of the spheres, the guided wave number hits the Bragg condition $\beta = \pi/d$, which ensures the exact relation $\mathbf{P}_{\mathrm{av}} = 0$, forcing the following conditions on the equivalent parameters:

$$\varepsilon_{\mathrm{eq}} = \varepsilon_0, \quad \mu_{\mathrm{eq}} = \mu_0 \pi^2/(k_0 d)^2. \quad (4)$$
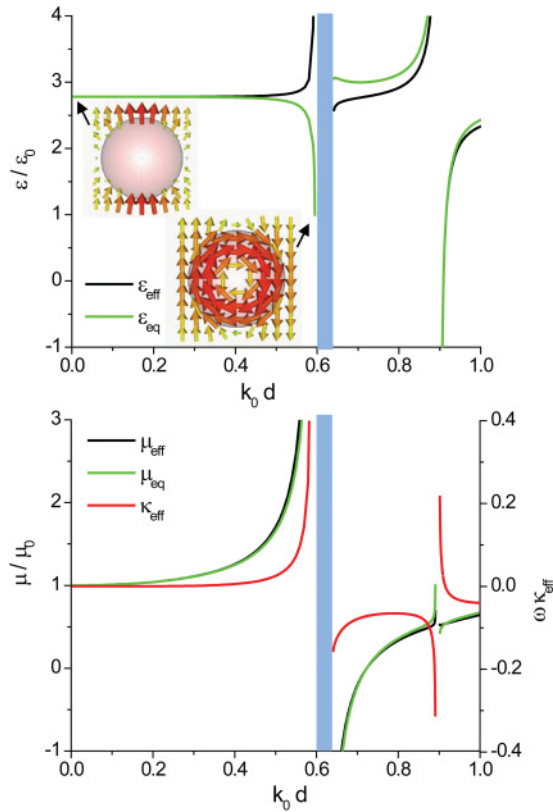
FIG. 2. (Color online) Effective and equivalent parameters (purely real, due to the absence of losses) for an array of dielectric spheres with $a/d = 0.45$, $\varepsilon = 120\varepsilon_0$, and $\mu = \mu_0$. The insets show the electric-field distribution in a unit cell in the E plane for $k_0 d = 0.1$ and $k_0 d = 0.59$ (brighter colors correspond to larger fields). Solid (dashed) lines correspond to real (imaginary) parts. The shadowed region indicates the array first (magnetic) band gap.

At the lower edge of the band gap, indeed the average electric polarization is identically zero, as confirmed by the field distributions shown in the inset of Fig. 2, calculated with full-wave numerical simulations inside one unit cell of the material at the two sample frequencies $k_0 d = 0.1$ and $k_0 d = 0.59$ (lower band-gap edge). At this second frequency, the unit cell indeed supports a strong magnetic resonance, which induces a rotating electric field inside the sphere, drastically different from its lower-frequency response, which is typically electric. The average electric polarization at $k_0 d = 0.59$ is evidently zero and, not surprisingly, the *equivalent* permittivity is exactly the same as the background, as predicted in Eq. (4). However, it is important to realize that the cancellation of electric response within the unit cell is not associated with an inherent lack of polarization of the sphere, but rather to the precise compensation of the strong electric polarization $(\varepsilon_{\text{eff}} - \varepsilon_0)\mathbf{E}_{\text{av}}$ by the magnetoelectric coupling $\kappa_{\text{eff}}\boldsymbol{\beta} \times \mathbf{H}_{\text{av}}$, as predicted by Eq. (2) in this eigenmodal regime. If we drop the eigenmodal assumption at this frequency and we let an additional electric field polarize the unit cell, without proportionally increasing $\mathbf{H}_{\text{av}}$, the pure magnetic response of the material would be modified, introducing a strong electric response, as correctly described in Eq. (2) and by ε_{eff}. It is evident that the isotropic model used in retrieval methods is not sufficient to describe this effect, causing antiresonant

artifacts in the equivalent parameters. The introduction of the magnetoelectric coupling in modeling the array ensures a causal Lorentzian response for both effective permittivity and permeability, accompanied by an analogous non-negligible resonance of κ_{eff}. Even for frequencies well below the first band gap, a significant divergence between equivalent and effective parameters is evident in Fig. 2, proving that these relevant magnetoelectric effects cannot be neglected even in the long-wavelength regime. The simple introduction of κ_{eff} in the metamaterial model completely restores the local nature of permittivity and permeability and allows describing the metamaterial response as a bulk, even very close to the inclusion resonance, where extreme parameters can be obtained.

Beyond the first band gap, a region of near-zero permeability is also obtained, for which inherent spatial dispersion effects were highlighted in Ref. 3. Also here the deviation
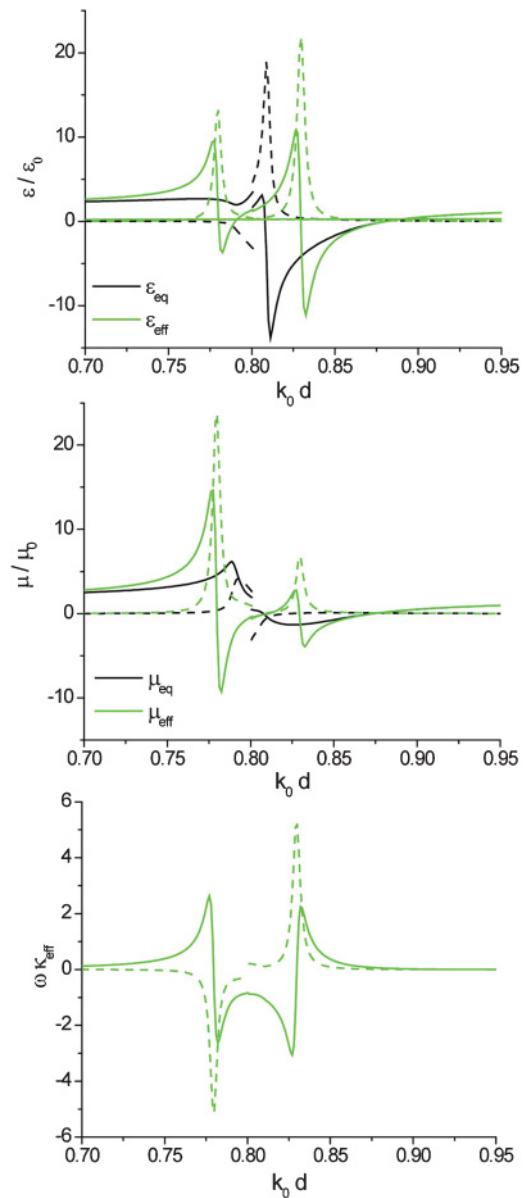

FIG. 3. (Color online) Effective and equivalent parameters for $a/d = 0.4$, $\varepsilon = (13.8 + i0.1)\varepsilon_0$, and $\mu = 11\mu_0$. Solid (dashed) lines correspond to real (imaginary) parts.

between equivalent and effective permittivity is rather large, due to the finite value of κ_{eff}, and $\varepsilon_{\mathrm{eq}}$ has a nonphysical negative dispersion. The introduction of κ_{eff} can evidently restore physically meaningful local constitutive parameters also in low-index metamaterials. Finally, at the second band gap $k_0 d \simeq 0.9$, dual considerations apply. Now $\mu_{\mathrm{eq}} = \mu_0$, and once again the effective parameters can considerably deviate from the equivalent ones.

As a second relevant example, consider an array of magnetodielectric spheres with $\varepsilon = (13.8 + i0.1)\varepsilon_0$, $\mu = 11\mu_0$, and $a/d = 0.4$. A similar geometry was considered in Ref. 7 to realize negative-index quasi-isotropic metamaterials, exploiting closely spaced electric and magnetic resonances achieved in commercially available magnetodielectric materials at microwave frequencies. Here, we also consider the presence of small Ohmic losses in the particles to verify the passivity conditions on the imaginary parts of the constitutive parameters in a NIM scenario.

The magnitude of the polarizability coefficients is shown in Fig. 1 (lighter green lines), showing a combined electric and magnetic resonance in the $k_0 d = 0.83$ range, which supports negative-index propagation in the band $k_0 d = 0.8$ to 0.9, as confirmed in Ref. 17. The presence of small losses helps to close the small band gap that would be present between the two resonances in the ideal lossless regime. Figure 3 compares effective and equivalent parameters extracted in this range (a more extended frequency range is reported in Ref. 17). The equivalent parameters (darker black lines) present all the nonphysical features typical of retrieved parameters in NIMs: antiresonant response, discontinuities, and blatant violation of passivity and causality requirements [Eq. (1)], showing that indeed these artifacts are inherently associated with the weak form of spatial dispersion highlighted here and associated with κ_{eff}. Indeed, the simple introduction of the magnetoelectric coefficient κ_{eff} in the constitutive model is seen to totally restore the continuity of the effective constitutive parameters, a strictly positive imaginary part, and clean Lorentzian resonances at the electric and magnetic closely spaced resonances of the array, as required by Kramers-Kronig

relations. The divergence between the equivalent and effective parameters in Fig. 3 is quite striking, and indeed it confirms the necessity to consider this simple correction, as in Eq. (2), which can totally restore the physical meaning of constitutive parameters even for negative index operation.

We have verified that the analytical results presented here, based on the analytical model introduced in Ref. 11, agree very well with full-wave simulations of these arrays, including the presence of higher-order multipoles, for arbitrary direction of propagation $\hat{\boldsymbol{\beta}}$ in the array, supporting a quasi-isotropic response. Reference 17 shows the comparison between the predicted value of eigenmodal $\beta = \omega\sqrt{\varepsilon_{\mathrm{eq}}\mu_{\mathrm{eq}}}$, as predicted by this theory using the polarizability coefficients to describe the wave interaction with the spheres, and full-wave simulations, ensuring that the dipolar approximation is very accurate and not at the basis of the artifacts pointed out here. In this paper, we have purposefully limited our analysis to purely isotropic inclusions and arrays, which allows us to isolate the inherent weak spatial dispersion effects at the basis of the presence of κ_{eff} in a proper homogenization model. More complex metamaterial geometries, including bianisotropic inclusions and asymmetric lattices, require considering a tensorial notation, but analogous second-order spatial-dispersion effects similarly arise and should be properly taken into account for the definition of physically meaningful homogenization parameters. It is relevant to stress that for larger values of $k_0 d$ and βd higher-order spatial-dispersion effects are generally present, making even the effective parameters defined here possibly nonlocal. The aim of this paper is to show that, even in the long-wavelength regime for which local metamaterial parameters are expected, magnetoelectric coupling stemming from the finite phase velocity along the lattice should be considered to obtain a physically meaningful macroscopic description of metamaterial arrays.

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[15]Notice that in the formulation introduced in Ref. 11 the magnetoelectric coupling coefficient $\kappa_{\mathrm{eff}} = \chi_{\mathrm{eff}}^0/\beta$.

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