Mie Scattering in the Macroscopic Response and the Photonic Bands of Metamaterials

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Herein, a general approach for the numerical calculation of the effective dielectric tensor of metamaterials is presented, and it is shown that the formalism may be used to study metamaterials beyond the long-wavelength limit. A system composed of high-refractive-index cylindrical inclusions is considered, and it is shown that the method reproduces the Mie resonant features and photonic band structure obtained from a multiple scattering approach, hence opening the possibility to study arbitrarily complex geometries for the design of resonance-based negative refractive metamaterials at optical wavelengths.

1. Introduction

Metamaterials are usually composed of periodic arrangements of micro- or nanostructures forming ordered patterns designed to control the propagation of light. They can display exotic optical properties with interesting applications, such as a negative refractive index which can be achieved exploiting underlying resonant features of the microstructure.1–5 Resonant behavior has been vastly studied in split-ring-resonator (SRR) structures, which are generally composed of metallic ring-like structures which give rise to a significant magnetic response when excited by an external inhomogeneous electric field. However, metamaterials based on SRRs are not well suited to visible frequencies, due to size limitations for their fabrication and high losses of the metallic components.6–8

Recently, high-refractive-index inclusions have been investigated as potential components for the fabrication of low-loss metamaterials displaying strong magnetic properties at optical frequencies.6–9 Moreover, the displacement current may increase with increasing permittivity, as in high-refractive-index particles the wavelength becomes shorter and can become comparable with the size of the particle, giving rise to Mie-like resonances. Thus, small high-index particles may display strong electric and magnetic Mie-like resonances. As these resonances are very sensitive to the dielectric environment and the corresponding fields are strongly enhanced at the surface, these particles have proven particularly useful in the development of biosensors based on several physical phenomena, as recently reviewed in the study by Krasnok et al.16 Moreover, these strong resonant fields are not confined to the surface of the particles, so they also yield large nonlinear effects within their small volumes, such as harmonic generation, sum and difference frequency generation, parametric down conversion, and others.17 In addition, low losses allow stronger external fields without damage. These high-index particles are for many applications superior to their plasmonic counterparts.

Structures with Mie-like resonances in the optical region can also be potentially used as resonators for the fabrication of low-loss negative refraction metamaterials. Negative refraction has been reported for microstructure geometries as simple as cylinders.12,18 Indeed, the number and frequencies of the resonant modes depend strongly on the size and geometry of the particles. Therefore, the generation and interference of such modes can be controlled by manipulating the composition, size, and shape of the particles, allowing a wide range of possibilities for the design of optical metamaterials. However, general methods to compute the effective response of metamaterials of arbitrary shape and composition are often limited to computationally expensive numerical approaches or approximations. If the wavelength of the incident light is long; compared with the microstructure of the metamaterial, its response can be described by an effective macroscopic dielectric function efficiently computed within the so-called “long wavelength limit.”19,20 Nevertheless, the description of Mie scattering lies by definition outside the validity range of the long wavelength limit generally used to study metamaterials. When the incident field varies in space on a length scale comparable with the microscale of the metamaterial, the effects of retardation and nonlocality become important. It has been shown that even when retardation effects are important, the system may be characterized by a macroscopic dielectric response,21 which must be described by a “nonlocal” tensor \( \epsilon(r, r', t, t') \). This nonlocal retarded response results in a frequency \( \omega \) and wavevector \( k \) dependence of the effective dielectric tensor in Fourier space, which leads to the constitutive equation \( D(\omega, k) = \epsilon(\omega, k) E(\omega, k) \).

In this article, we use a general efficient formalism for the numerical computation of the effective dielectric response of metamaterials at arbitrary wavelengths. We consider the case of a metamaterial composed by high-index dielectric cylinders.
and show that our approach can reproduce the analytical results obtained from Mie theory at wavelengths comparable with the size of the particle, thereby demonstrating that our method can be used to study metamaterials based on Mie resonances.

The structure of this article is the following. In Section 2, we present our theory for the calculation of the electromagnetic response of the metamaterial. First in Section 2.1, we develop an efficient computational method based on the calculation of the macroscopic dielectric response through a recursive procedure. This method is applicable to arbitrary materials and geometries. To interpret and test its results, in Section 2.2, we develop a multiple scattering approach applicable to an array of dielectric cylinders. Results on the dielectric response of high-index dielectric cylinders are presented in Section 3, compared with the results of the multiple scattering approach, and interpreted in terms of coupled Mie resonances. Finally, our conclusions are presented in Section 4.

2. Theoretical Methods

2.1. Macroscopic Response

We consider a binary metamaterial made up of two components, say, a host (A) and inclusions (B), each having well-defined dielectric functions \(\varepsilon_A\) and \(\varepsilon_B\). Its “microscopic” dielectric function is

\[
\varepsilon(r) = \begin{cases} 
\varepsilon_A, & r \in A \\
\varepsilon_B, & r \in B 
\end{cases}
\]

We abbreviate Equation (1) in terms of a structure function

\[
B(r) = \begin{cases} 
1, & r \in B \\
0, & r \notin B 
\end{cases}
\]

as \(\varepsilon(r) = \langle u - B(r)\rangle\), where \(u = 1/(1 - \varepsilon_A/\varepsilon_B)\) is known as the “spectral variable.” We write the inhomogeneous wave equation for the electric field \(E\) in the presence of an external current \(J_{\text{ext}}\) at a given frequency \(\omega\) as

\[
\hat{\nabla} \cdot \hat{\nabla} E = \left(\hat{\varepsilon} + \frac{1}{q^2} \nabla^2 \hat{\rho}^T\right) E = \frac{4\pi}{io\omega} J_{\text{ext}}
\]

which is obtained directly from Maxwell’s equations. Here, \(q \equiv \omega/c\) is the free-space wavenumber, and we treat the dielectric response as an operator instead of a simple function of position, as it acts on electric fields that may be described in real space, as functions of position, but also in reciprocal space, as functions of wavevectors.

In Equation (3), we have introduced a “wave operator” \(\hat{\nabla}\) written in terms of the Laplacian and the transverse projector \(\hat{\rho}^T = 1 - \hat{\rho}^L\), where 1 is the identity operator and \(\hat{\rho}^L\) is the longitudinal projector. We recall that, according to Helmholtz theorem, a vector field \(F\) may be written as the sum \(F = F^L + F^T\) of a longitudinal field \(F^L\) and a transverse field \(F^T\) which obeys \(\nabla \times F^T \equiv 0\) and \(\nabla \cdot F^T \equiv 0\). Thus, \(F^T\) may be written as the gradient of a scalar “potential” \(F^T = -\nabla \Phi\) which obeys \(\nabla^2 \Phi = -\nabla \cdot F^T = -\nabla \cdot F\). This may be solved formally as \(\Phi = -\nabla^2 \nabla \cdot F\), where \(\nabla^2\) is a convenient shorthand for the Green’s operator \(\hat{\nabla}\) of the Laplacian operator. \(\hat{\nabla}\) is represented in real space by an integral operator which acting on, say, \(\rho(r)\), yields the function \(\int d^3r' \rho(C(r, r'))\), where \(D = 1, 2, 3\) is the number of dimensions of space, and \(C(r, r')\) is a kernel which obeys Poisson’s equation \(\nabla^2 C(r, r') = \delta(r - r')\) with a unit point source. For example, for an unbounded space in 3D, \(C(r, r') = -1/(4\pi |r - r'|)\) is proportional to the Coulomb potential. In reciprocal space, the longitudinal projector can be written regardless of dimensionality as \(\hat{\rho}^L = \hat{k} k/k^2\), where \(k\) is the wavevector of magnitude \(k\).

We solve Equation (3) formally as

\[
E = \frac{4\pi}{io\omega} \hat{\nabla}^{-1} J_{\text{ext}}
\]

where \(\hat{\nabla}^{-1}\) is the inverse of the wave operator. Now we proceed to average this equation to obtain the “macroscopic” field \(E_M\) which we identify with a spatial average \(E\). To define what we mean precisely by this average, we consider a periodic system with Bravais lattice \(\{\mathbf{R}\} = \{\sum_i n_i \mathbf{d}_i\}\), where \(n_i\) are integers and \(d_i\) are primitive lattice vectors. According to Bloch’s theorem, any field \(F(r)\) may be written as a sum of Bloch waves

\[
F_k(r) = \sum_C F_C(k) \exp(i(k + G) \cdot r)
\]

with Fourier coefficients \(F_C(k)\) corresponding to wavevectors \(k + G\), where \(G\) is a vector of the reciprocal lattice defined by \(\exp(iG \cdot \mathbf{R}) = 1\) and \(k\) the Bloch’s vector which is a conserved quantity and may be chosen within the first Brillouin zone. Here, the long wavelength limit corresponds to \(k \ll \xi\). Notice that the field components with wavevectors \(k + G\) have spatial oscillations over relatively small distances of the order \(d_i\), except for the term \(G = 0\). Thus we define the spatial average \(F^\text{ext}(r)\) of the field \(F(r)\) through a truncation in reciprocal space, as \(F^\text{ext}(r) \equiv F_k \exp(i k \cdot r)\). Equivalently, in reciprocal space, we write \(F_{M,k}^\text{ext} = \delta_{0k} F_0\), which we write as the action of an averaging projector on the field \(F = \hat{\rho}^L F\). With this definition, we can represent the average operator in reciprocal space as the matrix

\[
\frac{\rho_{00}}{G_{00}} = \delta_{00} \delta_{\alpha0}
\]

As \(J_{\text{ext}}\) is an external current, it does not have spatial fluctuations due to the microstructure and \(J_{\text{ext}} = J_{\text{ext}}\). Thus, we average Equation (4) to obtain

\[
E_M = \frac{4\pi}{io\omega} \hat{\nabla}^{-1} J_{\text{ext}}
\]

where the “macroscopic” wave operator \(\hat{\nabla}^{-1}\) is given by \(\hat{\nabla}^{-1} = \hat{\rho}^L \hat{\nabla}^{-1} \hat{\rho}^L\), i.e., its inverse is the average of the inverse of the “microscopic” wave operator.

Substitution of the permittivity tensor in terms of the structure function, Equation (2), and the spectral variable \(u\) leads to the wave operator

\[
\hat{\nabla} \cdot \hat{\nabla} E = \hat{\varepsilon} (\mu - \hat{\nabla}^2) + \frac{1}{q^2} \nabla^2 \hat{\rho}^T
\]

which we rewrite as
\[ \mathcal{W} = \frac{c_A}{u} (u \mathbf{k}^{-1} - \hat{B}) \]

by introducing a “metric operator”

\[ \hat{g} = \left( 1 + \frac{\nabla^2}{q^2 e_A} \right)^{-1} \]

Inverting the wave operator and taking the average, we obtain

\[ \tilde{\mathcal{W}}^{-1} = \frac{\mu}{e_A} \hat{g}_{ab} (u - \hat{B} \hat{g})^{-1} \]

where we used the fact that the metric does not couple average to fluctuating fields.

Finally, we extract the macroscopic dielectric tensor from the corresponding wave operator

\[ \varepsilon_M(\omega, k) = \frac{1}{q} (k^2 \mathbf{I} - qk) + \mathcal{W}_M(\omega, k) \]

where we used the explicit transverse projector in reciprocal space for a plane wave with wavevector \( k \).

To compute the macroscopic dielectric tensor, we begin by calculating \((u - \hat{B} \hat{g})^{-1}\) in Equation (11). To that end, we first notice that the operator \( \hat{B} \hat{g} \) (Equation (2)) is Hermitian in the usual sense, even for lossy materials, as it depends exclusively on the geometry of the system, not on its composition. The operator \( \hat{g} \) would also be Hermitian if the response of medium A is dissipationless, i.e., if \( e_A \) is real. Nevertheless, the product \( \hat{B} \hat{g} \) is not Hermitian. We notice, however, that the product \( \hat{B} \hat{g} \) becomes Hermitian by redefining the internal product between two states, using \( \hat{g} \) as a metric tensor. Thus, we define the g-product of two states \( |\psi\rangle \) and \( |\phi\rangle \) as \((\phi|\psi)\), where

\[ (\phi|\psi) \equiv (\phi|\hat{g}|\psi) \] and \((\ldots|\ldots)\) is the usual Hermitian scalar product. With this definition, it is clear that

\[ (\phi|\hat{B} \hat{g} |\psi) = (\phi|\hat{g} \hat{B} |\psi) = (\psi|\hat{g} \hat{B} |\phi) = (\psi|\hat{B} \hat{g} |\phi) \]

so that \( \hat{B} \hat{g} \) is indeed Hermitian under the product \((\ldots|\ldots)\) and may borrow computational methods developed for quantum mechanical calculations.

We choose an initial state \( |0\rangle = b_0^{-1}|p\rangle \) where \(|p\rangle \) corresponds to a plane wave of frequency \( \omega \), wavevector \( k \), and polarization \( \varepsilon \), normalized as \(|p|p\rangle = 1 \) under the conventional internal product, and \( b_0 \) is chosen such that the state \( |0\rangle \) is \( g \)-normalized, \( \langle 0|0 \rangle = g_0 = \pm 1 \). Notice that as \( \hat{g} \) is not positive definite, we should allow for negative norms. We also define \(|-1\rangle = 0 \). Following the Haydock recursive scheme,[25] new states can be generated by repeatedly applying the Hermitian operator

\[ \hat{B} \hat{g} |n\rangle = b_{n+1}|n+1\rangle + a_n|n\rangle + b_n g_n b_{n-1}|n-1\rangle \]

where the real coefficients \( a_n, b_n, \) and \( g_n \) are obtained by imposing the orthonormality condition

\[ (n|m) = (n|\hat{g}|m) = g_n \delta_{nm} \] and \( g_n = \pm 1 \). Thus the operator \( \hat{B} \hat{g} \) has a tridiagonal representation in the basis \(|n\rangle\), which allows us to express the operator \((u - \hat{B} \hat{g})\) as

\[ (u - \hat{B} \hat{g}) = \begin{pmatrix} u - a_0 & -b_1 & 0 & \cdots \\ -b_1 & u - a_1 & -b_2 & 0 \\ 0 & -b_2 & u - a_2 & -b_3 & 0 \\ \vdots & \vdots & \ddots & \ddots & \ddots \end{pmatrix} \]

Finally, we have to invert and average the operator in Equation (17). We recall that the average (Equation (6)) is given in terms of a projection into our starting state \(|p\rangle\), i.e., the zeroth row and column element of the inverse operator which may be found for Equation (17) in the form of a continued fraction

\[ \hat{\varepsilon} \cdot (\mathcal{W}_M(\omega, k))^{-1} \hat{\varepsilon} = \frac{u e_A}{u - a_0} \frac{g_0 b_0^2}{u - a_0 - \frac{g_0 b_0^2}{u - a_1 - \frac{g_0 b_0^2}{u - a_2 - \frac{g_0 b_0^2}{\ldots}}}} \]

Choosing different independent polarizations \( \hat{\varepsilon} \) for the initial state \(|p\rangle\), one can compute all the independent projections of the inverse of the wave tensor. The result is then substituted in Equation (12) to obtain the fully retarded macroscopic dielectric tensor. Further details on the method and its implementation can be found in the study by Mochán et al.[21,26] We remark that the aforementioned procedure may be applied to two phase systems of arbitrary geometry and composition as long as one of them is dissipationless. The other one may well be dispersive and/or dissipative.

2.2. Scattering Approach

Multiple scattering from an ensemble of particles[27] is an old important problem whose study has had impact on many fields, such as atmospheric and acoustical[28],[29] biological and medical physics.[29] The theory of multiple scattering from cylinders was pioneered in the 1950s by Twersky.[10] In this section, we follow the study by Gagnon and Dubé[31] to compute the solution of the multiple scattering problem of a finite array of dielectric cylinders. Consider first a single infinitely long dielectric cylinder of radius \( R \) and refractive index \( n \), standing in vacuum with its axis aligned to the \( z \) axis. An incident field polarized on the \( x - y \) plane is applied. The magnetic field \( \mathbf{H} = (0, 0, \Phi) \) is taken parallel to the axis of the cylinder and satisfies scalar Helmholtz equations of the form

\[ \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial \Phi^\prime(r, \theta)}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \Phi^\prime(r, \theta)}{\partial \theta^2} \]

\[ + \frac{n^p(\omega)^2}{c^2} \Phi^\prime(r, \theta) = 0 \]

for each frequency \( \omega \), where \( n^p = n \) or 1 is the refractive index of the region \( \beta = I, O \), inside or outside of the cylinder, respectively.

Solutions of Equation (19) can be obtained as the products
$\Phi_l^0(x_l)\exp(\ii\theta_l)$, where $l = 0, \pm 1, \pm 2, \ldots$ and $\Phi_l^0$ solves the Bessel differential equation

$$\kappa_l^0 \frac{d^2}{dr^2} \left( \kappa_l^0 \frac{d\Phi_l^0}{dr} \right) + \left( (\kappa_l^0)^2 - \ell^2 \right) \Phi_l^0 = 0$$  \hspace{1cm} (20)

where $\kappa_l^0 = n_l^0 q r$. The general solution of Equation (20) inside (I) and outside (O) the cylinder can be written in terms of Bessel functions of the first and second kind $J_l$ and $Y_l$ as

$$\Phi^I(r, \theta) = \sum_{l} c_l J_l(q r) \exp(\ii\theta)$$

$$\Phi^O(r, \theta) = \sum_{l} [a_l J_l(q r) + b_l H_l(q r)] \exp(\ii\theta)$$  \hspace{1cm} (21)

$$\Phi^O(r, \theta) = \sum_{l} [a_l J_l(q r) + b_l H_l(q r)] \exp(\ii\theta)$$  \hspace{1cm} (22)

where we have chosen the outgoing Hankel functions $H_l = J_l + \ii Y_l$, as the scattered field. The coefficients $a_l$ describe the incident field and $b_l$ and $c_l$ are to be determined by the boundary conditions. These are the continuity of $H$ and the continuity of the component of the electric field $E = (\ii / \epsilon_0) \nabla \times H$ parallel to the interface, $E_o$, at $r = R$. As

$$E^I_0(r, \theta) = \frac{\ii}{n} \sum_{l} c_l J_l(q R \theta) \exp(\ii\theta)$$

$$E^O_0(r, \theta) = \sum_{l} [a_l J_l(q R \theta) + b_l H_l(q R \theta)] \exp(\ii\theta)$$  \hspace{1cm} (23)

$$E^O_0(r, \theta) = \sum_{l} [a_l J_l(q R \theta) + b_l H_l(q R \theta)] \exp(\ii\theta)$$  \hspace{1cm} (24)

where $J_l'$ and $H_l'$ denote the derivatives of the Bessel and Hankel functions with respect to their arguments; then

$$c_l J_l(n q R) = a_l J_l(q R) + b_l H_l(q R)$$

$$(1/n) c_l J_l(q R) = a_l J_l(q R) + b_l H_l(q R)$$  \hspace{1cm} (25)

which we solve for the scattering coefficients

$$s_l = \frac{b_l}{a_l} = \frac{J_l(q R)}{J_l'(q R)} - \frac{n J_l(q R) J_l'(q R)}{J_l(q R) H_l(q R) - H_l(q R) J_l'(q R)}$$

We consider now an array of $N \times N$ cylinders located at positions $R_m$. An incident plane wave traveling along the $x$-axis can be expressed in the frame of reference of the nth cylinder as

$$\Phi_m(r) = \exp(\ii k X_m) \sum_l \ii^n J_l(q R_m) \exp(\ii\theta_m)$$

where $r_m = (X_n, Y_n, Z_n)$ and $r - R_m$ are described by the polar coordinates $r_n$ and $\theta_m$. The Graf’s addition theorem allows us to rewrite a cylindrical function centered at $R_m'$ in a frame of reference centered at $R_m$: The wave scattered by cylinder $n'$ can be rewritten in the frame of reference of cylinder $n$ as

$$H_l(q R_m) \exp(\ii l' \theta_m) = \sum_l \exp \left( \ii (l - l') \phi_m \right)$$

$$\times H_{l - l'}(q R_m') J_l(q R_m) \exp(\ii\theta_m)$$

where $R_m' = R_m - R_m'$ is described by the polar coordinates $R_m'$ and $\phi_m$. Thus, the magnetic field in the interstices may be described in coordinates centered at the nth cylinder as the sum of the incident field, the nth scattered field, and the field scattered by all the other cylinders, as

$$\Phi_m(r) = \exp(\ii k X_m) \sum_l \ii^n J_l(q R_m) \exp(\ii\theta_m)$$

$$+ \sum_{n' \neq n} \sum_{l' \neq l} b_{n l} H_{l'}(q R_m) \exp(\ii(l - l') \phi_m)$$

$$\times H_{l - l'}(q R_m') J_l(q R_m) \exp(\ii\theta_m)$$  \hspace{1cm} (29)

From Equation (22) and (29), we identify the coefficients $a_{nl} as

$$a_{nl} = \exp(\ii k X_n) \delta_{ll'} + \sum_{n' \neq n} \sum_{l' \neq l} \exp \left( \ii (l - l') \phi_m \right)$$

$$\times H_{l - l'}(q R_m') b_{n l'}$$

Introducing the scattering coefficients $s_{nl}$ of each cylinder as in (26) yields

$$b_{nl} = s_{nl} \sum_{n' \neq n} \sum_{l' \neq l} \exp \left( \ii (l - l') \phi_m \right)$$

$$\times H_{l - l'}(q R_m') b_{n l'} = s_{nl} \exp(\ii k X_n)$$

which can be summarized into a system of coupled equations

$$Tb = a$$  \hspace{1cm} (32)

where $a = \{s_{nl} \exp(\ii k X_n)\}$ describes the incident field, $b = \{b_{nl}\}$ describes the scattered field in the interstitial region, to be obtained, and

$$T_{nl'} = \delta_{nl} \delta_{ll'} - \delta_{nl}$$

$$\times \exp \left( \ii (l - l') \phi_m \right) H_{l - l'}(q R_m') s_{nl}$$  \hspace{1cm} (33)

3. Results

We consider a metamaterial made of a square lattice of identical infinitely long dielectric cylinders of radius $R$ and refractive index $n$ set in vacuum, with a lattice constant $a$. Using an efficient computational implementation of the numerical approach presented in Section 2.1, we calculate the macroscopic dielectric function of the metamaterial as a function of frequency $\omega$ and wavevector $k$. We compare our results with the analytical solution obtained in Section 2.2 for a finite array of $N \times N$ cylinders.

We first consider thin weakly interacting cylinders with radius $R = 0.1a$ and refractive index $n = 10$. Numerical calculations were carried out using a 2D $601 \times 601$ grid to discretize the unit cell and carried out the recursive calculation using 450 Haydock coefficients. For the analytical case, we considered a large finite array of $511 \times 511$ cylinders and a maximum value of the orbital number $l = 1$. We checked the convergence of the results by repeating the calculations with larger grids, more Haydock coefficients, and larger angular momenta. Figure 1a shows the results for the transverse component of the macroscopic dielectric tensor of the metamaterial $\varepsilon_{TM} = \varepsilon_T^0$, obtained through
The recursive numerical approach. The results are shown as a function of the frequency, characterized by \( nqR \), for a wavevector along the \( x \) direction slightly larger than the vacuum wavevector \( k = 1.01q \).

Three prominent resonant features are observed in both panels of Figure 1 at low energies. The lowest energy resonance corresponds to a magnetic dipole arising from the term \( l = 0 \), as it lies close to that of an isolated cylinder occurring at the first zero of the Bessel function around \( nqR = 2.4 \). The second resonance around \( nqR \approx \pi \) emerges from the fulfillment of Bragg’s diffraction condition for \( 2a = 2\pi/q \). A third resonance close to \( nqR \approx 3.8 \) is originated by the term \( l = 1 \). This resonance is strongly enhanced through the interaction between cylinders. The additional peaks in the macroscopic response are due to resonances caused by multiple reflections in the interstitial regions. We have verified that they are not due to numerical noise, but they disappear when a very small artificial dissipation is added to the interstitial dielectric function, whereas the large peaks are robust. Notice however that the scattering coefficients

\[ b_0 \] resonate at higher energies than the macroscopic dielectric function. The reason for this discrepancy is that the transverse normal modes of the system are not actually given by the poles of the dielectric response but by the poles of the electromagnetic Green’s function \( G \).

**Figure 2** shows the imaginary part of the electromagnetic Green’s function \( G = (\varepsilon_M^\prime - k^2/q^2)^{-1} \). For display purposes, we introduced a small artificial broadening parameter \( \eta = 0.001 \) and replaced \( \text{Im}G = \eta/((\varepsilon_M^\prime - k^2/q^2)^2 + \eta^2) \). The figure also shows the squared magnitude of the scattering amplitude \( b_0 = \sum b^{(i)}_0 \) for \( k \approx q \) as a function of \( nqR \). Here, the peaks of the Green’s function coincide with the peaks of the scattering coefficient as the poles of the Green’s function and of the scattered coefficient correspond both to the normal modes of the system, for which one may have a finite field and finite scattered amplitudes without an external excitation. The widths \( \Delta\omega \) of the peaks in Figure 2a are due to the artificial broadening parameter and are of the order of \( \eta/(d\varepsilon_M^\prime/d\omega) \), evaluated at resonance. The widths of the peaks in Figure 2b are due to radiation losses, as the corresponding system is large but finite.

Having verified the mutual consistency of both computational approaches when applied to the calculation of the normal modes of a system, we now consider a more interesting and realistic case. We consider a metamaterial composed of strongly interacting cylinders with a larger radius \( R = 0.35a \) and a large but realistic refraction index \( n = 4 \) (similar to that of Si). To identify exotic behavior such as negative refraction, we have to examine the dispersion relation of the normal modes of the system.
and explore their group velocity.\textsuperscript{22,33} Thus, we calculated the macroscopic dielectric function and obtained the Green’s function of the metamaterial as a function of both frequency \(\omega\) and wavevector \(k\).

For the numerical calculations, we used a 2D 201 \(\times\) 201 grid and sets of 300 Haydock coefficients. A very small artificial dissipative term 0.01\(i\) has been added to the vacuum dielectric constant to improve convergence. The results are shown in Figure 3. The scattering amplitude obtained from the scattering approach, calculated for an array of 201 \(\times\) 201 cylinders, considering a maximum value of the orbital number \(l = 2\), is shown for comparison.

The upper panel of Figure 3 shows the imaginary part of Green’s function \(G^\prime = \eta/(\epsilon_T^M - k^2/q^2 + \eta^2)\), where \(\epsilon_T^M\) was obtained numerically through Haydock’s recursion and the results have been smoothed with a dissipation factor \(\eta = 0.2\) for better visualization. The lower panel shows the absolute value square of the scattering amplitude \(b_0 = \sum_l b_0^l\) smoothed as \(\eta/(1/|b_0|^2 + \eta^2)\) with a dissipation factor \(\eta = 0.1\).

Several bands may be identified in Figure 3, showing a very good agreement between the dispersion relations obtained through our two approaches. Regions of negative dispersion, i.e., for which the frequency of the resonances decreases as the wavevector increases, yielding a negative group velocity, are clearly observed for the second, fourth, and fifth bands and for large wavevectors before the first Brillouin zone boundary at \(ka = \pi\). The bands originate from the combination of isolated cylinder resonances of different values of \(l\), as different values of \(l\) participate in each band due to the relatively strong interaction between neighboring thick cylinders. This can be confirmed through Figure 4, which shows the absolute value of the individual scattering coefficients \(b_0^l\) for \(l = 0, 1, 2\) as a function of \(nqR\) for a) a single cylinder, b) an array of 11 \(\times\) 11 cylinders, and c) an array of 201 \(\times\) 201, for a wavevector \(k = q\). The coefficients corresponding to an isolated cylinder display one broad peak each around \(q = 2.4, 4.8\) and 5.1 for \(l = 0, 1, 2\) respectively, which are close to the first zeroes of the corresponding Bessel function. For an array of 11 \(\times\) 11 cylinders,

![Figure 3](image-url)  
**Figure 3.** a) Imaginary part of the Green’s function \((\epsilon_T^M - k^2/q^2)^{-1}\) for a system composed of dielectric cylinders of radius \(R = 0.35a\) and refractive index \(n = 4\), obtained numerically through the macroscopic response using the package “Photonic”. b) Magnitude of the scattered amplitude \(b_0\) normalized as \(\eta/(1/|b_0|^2 + \eta^2)\) with a dissipation factor \(\eta = 0.1\).

![Figure 4](image-url)  
**Figure 4.** Absolute value of the scattering coefficients \(|b_0^l|\) for an a) isolated cylinder, b) array of 11 \(\times\) 11 cylinders, and c) array of 201 \(\times\) 201 cylinders, as a function of \(nqR\).
these peaks appear distorted and shift to be finally merged for a larger array of $201 \times 201$ in which case all $l$ contributions resonate at the same frequencies.

4. Conclusions

We presented two schemes to calculate the electromagnetic properties of a metamaterial made of a simple lattice of cylinders: a numerical method based on a recursive calculation of the macroscopic dielectric tensor which may be easily generalizable to arbitrary geometries and materials and a multiple scattering approach for cylindrical geometries which allowed us a simple interpretation of the results in terms of the excitation of Mie resonances. We applied these methods to investigate the response of a system made up of cylinders of a high index of refraction. The comparison between the results of both methods is not direct, as in one case, we obtain a macroscopic response and in the other, we obtain scattering amplitudes. Nevertheless, we showed that the poles of the macroscopic Green's function obtained by our numerical method coincide with those of the scattering coefficients, and both can be interpreted as the excitation of the normal modes of the system. For a system of thin cylinders with a very high index of refraction and a relatively small coupling, we identified the nature of each mode. We found modes arising from the Mie resonances of individual cylinders and a mode arising from the Bragg coherent multiple scattering. For larger cylinders, the interaction yields a coupling between resonances with different angular momenta. By varying the frequency and wavevector independently, we computed the dispersion relation of the normal modes. The photonic band structure obtained using both methods is in very good agreement and reveals regions of negative dispersion. Thus, through comparison with an ad-hoc model, we showed that our macroscopic approach based on Haydock's recursion and its implementation in the "Photonic" package is an efficient procedure for obtaining the optical properties of metamaterials made up of cylinders with high index of refraction incorporating resonances which cannot be explored within the long wavelength limit. Furthermore, as it can be readily generalized to arbitrary geometries and materials, we believe it will prove to be a useful tool for the design of artificial materials with a richer geometry that might yield novel properties.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

homogenization, metamaterials, Mie resonances, nonlocal optics

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