Electromagnetic coupling and array packing induce exchange of dominance on complex modes in 3D periodic arrays of spheres with large permittivity

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We investigate the effect on wave propagation of array packing and electromagnetic coupling between spheres in a three-dimensional (3D) lattice of microspheres with large permittivity that exhibit strong magnetic polarizability. We report on the complex wavenumber of Bloch waves in the lattice when each sphere is assumed to possess both electric and magnetic dipoles and full electromagnetic coupling is accounted for. While for small material-filling fractions we always determine one dominant mode with low attenuation constant, the same does not happen for large filling fractions, when electromagnetic coupling is included. In the latter case we peculiarly observe two dominant modes with low attenuation constant, dominant in different frequency ranges. The filling fraction threshold for which two dominant modes appear varies for different metamaterial constituents, as proven by considering spheres made by either titanium dioxide or lead telluride. As further confirmation of our findings, we retrieve the complex propagation constant of the dominant mode(s) via a field fitting procedure employing two sets of waves (direct and reflected) pertaining to two distinct modes, strengthening the presence of the two distinct dominant modes for increasing filling fractions. However, given that one mode only, with transverse polarization, at any given frequency, is dominant and able to propagate inside the lattice, we are able to accurately treat the metamaterial that is known to exhibit artificial magnetism as a homogeneous material with effective parameters, such as the refractive index. Results clearly show that the account of both electric and magnetic scattering processes in evaluating all electromagnetic intersphere couplings is essential for a proper description of the electromagnetic propagation in lattices. © 2016 Optical Society of America

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1. INTRODUCTION

The characterization and understanding of wave propagation in composite materials such as arrays of particles periodic in three dimensions are of great importance for improving the design of metamaterials and as such have been greatly discussed in the literature [1–20]. In particular, collective resonances in three-dimensional (3D) periodic metamaterials can be tailored to create artificial dielectric and magnetic behavior, narrowband absorption, as well as backward and quasi-dark modes.

Starting from [15], it has been pointed out that electromagnetic coupling and spatial dispersion might affect strongly the metamaterial response and as such should be included in the characterization of wave propagation in 3D lattices. This has been further remarked on in [16,17,21], and very recently in [19,20,22–24]. These contributions notwithstanding, mode propagation with complex wavenumber has seldom been analyzed when electromagnetic coupling is accounted for between microspheres and array packing is varied. Thus, the aim of this paper is to characterize the Bloch modes with complex wavenumber in 3D periodic lattices made of arrayed particles with spherical shape that are modeled here as possessing both electric and magnetic dipole moments, a technique referred to as the dual (electric and magnetic) dipole approximation (DDA) introduced in [25]. We discuss here the effect on the Bloch wavenumber when electromagnetic coupling, due to array packing, is varied, showing that for large filling fractions some kind of “mode degeneracy” (intended here as exchange of dominance) takes place. The occurrence of this effect highlights the
importance of accounting for dynamic electromagnetic coupling and both electric and magnetic dipoles in mode characterization. Indeed, we show that the DDA is a good approximation when the two dipolar terms (or any of them) dominate the scattered-field multipole expansion. The numerical procedure used in this paper for evaluating the complex zeros of the dispersion relation uses the Ewald representation for the dyadic periodic Green’s functions (GFs) to represent the field in 3D periodic arrays (as discussed in [25] and references therein) because it provides us with analytic continuation to the complex wavenumber space and with series with Gaussian convergence.

The structure of the paper is as follows. We discuss in Section 2 the dipolar response of a single spherical particle through its electric and magnetic polarizabilities. In Section 3, we first summarize the formulation recently reported in [25] and then employ it to characterize modal (real and complex) wavenumbers for transverse polarization (with respect to the mode traveling direction) in two examples of composite metamaterials: one made of a 3D lattice of nonmagnetic titanium dioxide (TiO$_2$) microspheres, and the other made of nonmagnetic lead telluride (PbTe) microspheres. We recall here that in [25], we have shown only dispersion diagrams relative to modal branches here marked as dominant, as it will be discussed in later sections. We report here a more complex dynamics depending on the filling fraction. Then, in Section 4, we consider various arrays by changing the filling fraction, with emphasis on electromagnetic coupling effects. This analysis was not performed before, to the authors’ knowledge, and as such may lead to a better understanding of modal analysis in periodic systems and of magneto-dielectric coupling varying the filling factor. We discuss in Section 5 the dominance properties of each Bloch mode in the 3D lattice through the field fitting procedure introduced in [11]. We also show that the dominance analysis allows for the retrieval of the dispersion diagrams and this is shown to be in good agreement with the one obtained via DDA formulation discussed in the previous sections. Finally, since one mode only, with transverse polarization, is dominant and able to propagate inside the lattice at any given frequency, we treat in Section 6 the metamaterial as a homogeneous material with effective parameters, showing the refractive index computed via DDA compared to the same result retrieved via full-wave simulations.

2. SPHERE RESONANCES: ELECTRIC AND MAGNETIC DIPOLE POLARIZABILITIES

Consider the 3D lattice (i.e., a 3D periodic array) of spheres with relative permittivity $\varepsilon_r$ and radius $r$ shown in Fig. 1.

We consider TiO$_2$ and PbTe microspheres to obtain interesting wave propagation in the millimeter and mid-infrared frequency ranges, respectively. Both materials possess large permittivity values, which enable the rise of artificial magnetism via the excitation of a Mie resonance with circulating electric fields. The frequency dependence of the TiO$_2$ permittivity $\varepsilon_r = \varepsilon'' + i\varepsilon''$ is assumed as in [26,27], where $\varepsilon'' = 3.33f^2 + 92.34$ and $\varepsilon'' = 0.28f^2 + 7.64f - 1.54$, with $f$ being the frequency in terahertz. The frequency dependence of the PbTe permittivity is taken from [28]. Note that in the case of TiO$_2$ the relative permittivity is in the range of 93–94 for the real part and of 0–2.35 for the imaginary part [26,27], whereas for PbTe the relative permittivity is in the range of 28–34 for the real part and of 0.05–0.19 for the imaginary part.

The normalized (and unitless) sphere electric and magnetic dipole moments under the dipole approximation for the zeroth reference sphere at position $r_0$ are given by [29,30]

$$p_0 = \alpha_{ee} \cdot E^{loc}(r_0),$$

$$m_0 = \alpha_{mm} \cdot H^{loc}(r_0),$$

(1)

where $E^{loc}(r_0)$ and $H^{loc}(r_0)$ are the local electric and magnetic fields acting on the reference sphere at $r_0$, and $\alpha_{ee} = \alpha_{ee} I$ and $\alpha_{mm} = \alpha_{mm} I$ are the sphere electric and magnetic polarizability tensors, with $\alpha_{ee}$ and $\alpha_{mm}$ being the isotropic electric and magnetic polarizabilities and $I$ being the unit dyad. These polarizabilities are readily connected to the Mie dipolar coefficients for spherical scatterers $a_1$ and $b_1$ by $\alpha_{ee} = 6\pi i k r a_1/k_0^2$ and $\alpha_{mm} = 6\pi i k r b_1/k_0^2$, with $k_0 = \omega_0/\varepsilon_0^0$ and $k_0$ being the free-space wavenumber, using the notation in [31], to which the reader is referred for more information (see also [3,11,25]). The normalized (and unillss) sphere’s electric (normalized to the host absolute permittivity $\varepsilon_0$) and to the sphere volume $V_0$.

![Fig. 1. Sketch of a 3D lattice of spheres with radius r and relative permittivity $\varepsilon_r$ embedded in a homogeneous host with relative permittivity $\varepsilon_h$. The periodicities along the x, y, and z directions are denoted as a, b, and c, respectively.](image)

![Fig. 2. Electric ($\alpha_{ee}$, normalized to the host absolute permittivity $\varepsilon_0$) and to the sphere volume $V_0$) and magnetic ($\alpha_{mm}$, normalized to the sphere volume $V_0$) polarizabilities versus frequency for (a) a TiO$_2$ sphere with radius $r = 52$ μm and (b) a PbTe sphere with radius $r = 1$ μm, embedded in free space with relative permittivity $\varepsilon_r = 1$.](image)
$V_r = \frac{4\pi a^3}{3}$ and magnetic (normalized to the sphere volume $V_r$) polarizabilities are shown in Fig. 2 for the two materials considered. The magnetic resonance is sharper because of loop-like circulating displacement currents (equivalent to a magnetic dipole) of smaller scattering losses than electric dipoles.

For increasing frequency, we observe first a magnetic dipole resonance (around 300 GHz for a TiO$_2$ sphere and around 26 THz for a PbTe sphere) with residual electric contributions, followed by an electric dipole resonance (around 420 GHz for a TiO$_2$ sphere and around 36 THz for a PbTe sphere) with residual magnetic contributions, as expected from a spherical resonator with large dielectric permittivity, as also shown in [25,32], for example. We also note that in the case of TiO$_2$ spheres, the magnetic resonance is much stronger than the electric one, whereas in the case of PbTe spheres the two resonances have comparable strengths, though the magnetic one is still slightly stronger. This point can also be inferred by the Mie coefficients results reported in [25]. In the following sections we will mainly focus on the frequency region pertaining to the magnetic resonance because we are interested in artificial magnetism (i.e., macroscopic effective relative permeability different than unity) and interesting modal propagation features appear when electromagnetic coupling is included. We further stress that magnetic and electric contributions are present in a wide spectrum, as shown by the polarizability plot in Fig. 2; thus, electromagnetic coupling can happen in a wide frequency spectrum.

3. MODAL ANALYSIS: PROPAGATION IN A 3D PERIODIC LATTICE

We are interested in understanding how the electromagnetic coupling among spheres and the array packing affect mode propagation in 3D lattices. To this aim, we employ modal analysis (MA) to calculate the modes with either real or complex wavenumber using the above-mentioned dual (electric and magnetic) dipole approximation (MA-DDA). The dipolar approximations are accurate when any of the fields by the electric or magnetic dipolar terms dominate the scattered-field multipole expansion. Under these conditions, limiting the resonator interactions to only electric and magnetic dipole-dipole terms is a satisfactory approximation for a large class of problems involving collections of nanoresonators or microresonators [33]. The results obtained with the DDA are then compared with those obtained assuming each sphere is modeled simply as a single magnetic dipole, a technique named single dipole approximation (SDA), as in [11].

We analyze two structures, namely, a 3D lattice of TiO$_2$ spheres (Structure A) and PbTe spheres (Structure B) whose normalized electric and magnetic polarizabilities are shown in Fig. 2. We consider cubic lattices with periodicities of $a = b = c$ varying as in Tables 1 and 2, where for each structure we analyze various cases of filling fractions $ff = V_r/a^3$, depicted with Roman numerals I–VI for Structure A and I–III for Structure B.

### Table 1. Parameters of the 3D Lattices of TiO$_2$ Spheres Considered in This Paper

<table>
<thead>
<tr>
<th>Structure</th>
<th>Material</th>
<th>$a$ [µm]</th>
<th>$d$ [µm]</th>
<th>$ff$ [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>A-I</td>
<td>TiO$_2$</td>
<td>52.0</td>
<td>136.0</td>
<td>23.4</td>
</tr>
<tr>
<td>A-II</td>
<td>TiO$_2$</td>
<td>52.0</td>
<td>126.0</td>
<td>29.4</td>
</tr>
<tr>
<td>A-III</td>
<td>TiO$_2$</td>
<td>52.0</td>
<td>120.0</td>
<td>34.1</td>
</tr>
<tr>
<td>A-IV</td>
<td>TiO$_2$</td>
<td>52.0</td>
<td>118.0</td>
<td>35.8</td>
</tr>
<tr>
<td>A-V</td>
<td>TiO$_2$</td>
<td>52.0</td>
<td>116.0</td>
<td>37.7</td>
</tr>
<tr>
<td>A-VI</td>
<td>TiO$_2$</td>
<td>52.0</td>
<td>106.0</td>
<td>49.4</td>
</tr>
</tbody>
</table>

### Table 2. Parameters of the 3D Lattices of PbTe Spheres Considered in This Paper

<table>
<thead>
<tr>
<th>Structure</th>
<th>Material</th>
<th>$a$ [µm]</th>
<th>$d$ [µm]</th>
<th>$ff$ [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>B-I</td>
<td>PbTe</td>
<td>1.0</td>
<td>4.0</td>
<td>6.5</td>
</tr>
<tr>
<td>B-II</td>
<td>PbTe</td>
<td>1.0</td>
<td>3.0</td>
<td>15.5</td>
</tr>
<tr>
<td>B-III</td>
<td>PbTe</td>
<td>1.0</td>
<td>2.5</td>
<td>26.8</td>
</tr>
</tbody>
</table>

A. Analytical Procedure for Mode Analysis Using DDA and SDA

The employed analytical procedure uses periodic GFs based on the Ewald representation to compute the modal wavenumbers in 3D periodic arrays, including the case of complex modal wavenumbers, and is here briefly summarized (we refer the reader to [3,11,25] for more details).

Consider the 3D periodic array of spheres in Fig. 1, with each sphere described by dipole-like electric and magnetic polarizabilities in Eq. (1). Spheres are placed at positions $r_n = r_0 + d_n$, where $d_n = n_1a\hat{x} + n_2b\hat{y} + n_3c\hat{z}$; $n = (n_1, n_2, n_3)$ is a triple index with $n_1, n_2, n_3 = 0, \pm 1, \pm 2, \ldots$; $r_0 = x_0\hat{x} + y_0\hat{y} + z_0\hat{z}$ denotes a reference sphere location; and $a$, $b$, and $c$ are the periodicities along the $x$, $y$, and $z$ directions, respectively [1,3,11,25]. Suppose that the 3D array supports a Bloch mode with wave vector $k_0 = k_x\hat{x} + k_y\hat{y} + k_z\hat{z}$ such that the $n$th dipole (at $r_n$) is related to the zeroth one (at $r_0$) by

$$p_n = p_0 \exp(i k_0 \cdot d_n),$$

$$m_n = m_0 \exp(i k_0 \cdot d_n),$$

with $p_0$ and $m_0$ as in Eq. (1). Note that since we are interested in computing the modes excitable in the 3D lattice, we consider self-sustaining solutions, i.e., vanishing external field. Next, according to [25], we express the local fields $E_{loc}(r_0)$ and $H_{loc}(r_0)$ as the field generated by all spheres except the zeroth one,

$$E_{loc}(r_0) = \sum_{n \neq 0} \mathbf{G}_{EE}^{sc}(r_0, r_n, k_0) \cdot p_n + \mathbf{G}_{EM}^{sc}(r_0, r_n, k_0) \cdot m_n,$$

$$H_{loc}(r_0) = \sum_{n \neq 0} \mathbf{G}_{EF}^{sc}(r_0, r_n, k_0) \cdot p_n + \mathbf{G}_{HM}^{sc}(r_0, r_n, k_0) \cdot m_n,$$

where we account for both electric and magnetic dipolar scattering. Indeed, the terms $\mathbf{G}^{sc}$ are the regularized $3 \times 3$ dyadic GFs for electric–electric, electric–magnetic, magnetic–electric, and magnetic–magnetic interactions, where the field contribution from the zeroth resonator onto itself has been removed (more details can be found in [25]). One can observe from Eq. (3) that electromagnetic coupling is an effect that induces a modification of the local electric field from a magnetic dipole or a modification of the local magnetic field from an electric dipole. This usually is a negligible effect in the presence of only electric or magnetic resonances. This contribution is, however, important when magnetic and electric resonances are present, as in the system analyzed here, or in frequency regions away from such resonances. Then, following the procedure detailed
in [25], one obtains a linear system from which for any fixed frequency one can compute the modal Bloch wavenumbers $k_B$ by solving

$$\det(\|\|^{-1} - \Lambda(k_0)) = 0$$

(4)

for complex $k_B$. Here,

$$\alpha = \begin{bmatrix} \alpha_x & 0 \\ 0 & \alpha_{mm} \end{bmatrix}$$

(5)

and

$$\Lambda(k_0) = \begin{bmatrix} G_{EM}(r_0, r_0, k_B) & G_{EM}(r_0, r_0, k_B) \\ G_{HP}(r_0, r_0, k_B) & G_{HP}(r_0, r_0, k_B) \end{bmatrix}.$$  

(6)

We employ the Ewald method for fast computation of the regularized (i.e., without the zeroth term) GFs in Eq. (6) because it provides us with the analytic continuation of $k_B$ into the complex wavenumber space and it is made of just of a few terms exhibiting rapid Gaussian convergence. The dyadic expressions for the required spatial and spectral terms making up the Ewald representation have been reported in [25] for a 3D periodic array. The wavenumber $k_B = k_z \hat{z}$ for transversely polarized (electric field polarized along $x$) modes traveling along $z$ can also be computed by solving the scalar equation [19,23]

$$[\alpha_x^{-1} - G_{EM,xx}(r_0, r_0, k_B)][\alpha_{mn}^{-1} - G_{HM,yy}(r_0, r_0, k_B)] - G_{EM,xy} G_{HP,xy} = 0,$$

(7)

where $G_{EM,xx}$ is the $xx$ component of $G_{EM}$, $G_{EM,yy}$ is the $yy$ component of $G_{EM}$, $G_{EM,xy}$ is the $xy$ component of $G_{EM}$, and $G_{HP,xy}$ is the $xy$ component of $G_{HP}$. One can thus see that in view of Eq. (6) or Eq. (7) electromagnetic coupling between magnetic and electric fields due to the terms $G_{EM}$ and $G_{HP}$ can strongly affect the dispersion properties of the 3D metamaterial under analysis. When neglecting electromagnetic coupling, i.e., $G_{EM,xy} = G_{HP,xy} = 0$, Eq. (7) simplifies to

$$[\alpha_x^{-1} - G_{EM,xx}(r_0, r_0, k_B)][\alpha_{mn}^{-1} - G_{HM,yy}(r_0, r_0, k_B)] = 0,$$

(8)

where $\alpha_x - G_{EM,xx}(r_0, r_0, k_B) = 0$ provides the $k_B$ solution relative to single (electric) dipole approximation (MA-SDA-E) [3], whereas $\alpha_{mn}^{-1} - G_{HM,yy}(r_0, r_0, k_B) = 0$ provides the $k_B$ solution relative to single (magnetic) dipole approximation (MA-SDA-M) [11], respectively. Around the magnetic resonance, as in the plots in Section 4, we will show the modal wavenumber $k_B$ solution of $\alpha_{mn}^{-1} - G_{HM,yy}(r_0, r_0, k_B) = 0$.

Since Eq. (7) contains the effect of electromagnetic coupling, whereas Eq. (8) does not include such an effect, we will make use of both equations in the next subsection to better highlight when it is important to consider magneto-dielectric coupling.

### B. Dispersion Diagram for Transverse Polarization (T-pol) for Structures A-VI and B-II

We consider that modes propagate in the $z$ direction with Bloch wavenumber $k_B = k_z \hat{z}$ with complex $k_z = \beta_z + i\alpha_z$, where $\beta_z$ is the modal propagation constant in the first Brillouin zone $-1 < \beta_z a / \pi < 1$ and $\alpha_z$ is the modal attenuation constant. Since we consider waves in the lattice with transverse polarization, dipole moments are transversely polarized (T-pol) with respect to the mode direction of propagation. For modes propagating in the positive $z$ direction, only modes with $\alpha_z > 0$ satisfy the physical decay condition at $z = +\infty$. Modes are classified as forward when $\beta_z \alpha_z > 0$ and backward when $\beta_z \alpha_z < 0$ [3].

We thus show in Fig. 3 the wavenumber dispersion diagram relating real and imaginary parts of the modal wavenumbers of three modes to frequency. We show results pertaining to both Structure A-VI and Structure B-II. We observe the presence of three modes with attenuation not exceeding $\alpha_z a / \pi < 2$, two forward (Modes 1 and 2) and one backward (Mode 3), for both structures. Other modes with transverse polarization with normalized attenuation constant $\alpha_z$ larger than 2 are present, though not reported as highly attenuated. It is also important to note that the backward mode (Mode 3) never has a low attenuation constant, i.e., it has $\alpha_z a / \pi > 1$ in the shown frequency range.

In the case of Structure B-II, we observe that one mode only (Mode 1) presents $\alpha_z$ lower than any of the other modes in the entire frequency range analyzed. The same does not happen for Structure A-VI, where we note that two modes have the lowest $\alpha_z$ in two different frequency bands: Mode 1 between 200 and 300 GHz and Mode 2 between 300 and 350 GHz. This plot makes us infer that under certain conditions imposed by the modal attenuation constants, modes can exchange dominance, and this is better investigated in the next section.

### 4. EFFECT OF ARRAY PACKING AND ELECTROMAGNETIC COUPLING ON MODES WITH COMPLEX WAVENUMBER

We investigate in this section the effect of array packing (i.e., the filling fraction) and electromagnetic coupling on modes with complex wavenumber supported by the 3D lattice and on
possible intersection of dispersion curves. We thus show in Fig. 4 the trajectories of modal wavenumbers in the complex plane $\beta_z, \alpha_z$ for Structures A-I to A-VI, calculated with MA-DDA. Arrows indicate direction of increasing frequency. For structure A mode degeneracy takes place between Mode 1 and Mode 2 for filling factors larger than 35%, i.e., for cases A-IV, A-V, and A-VI. Note that Modes 1 and 2 are forward since they are in the first and third complex quadrants, whereas Mode 3 is backward. The frequency range is the same as the one analyzed in Fig. 3.

Fig. 4. Trajectories of modal wavenumbers in the complex plane $\beta_z, \alpha_z$ for Structures A-I to A-VI, calculated with MA-DDA. Arrows indicate direction of increasing frequency. For structure A mode degeneracy takes place between Mode 1 and Mode 2 for filling factors larger than 35%, i.e., for cases A-IV, A-V, and A-VI. Note that Modes 1 and 2 are forward since they are in the first and third complex quadrants, whereas Mode 3 is backward. The frequency range is the same as the one analyzed in Fig. 3.

The investigation of Bloch modes in Structures B-I to B-III is shown in Fig. 7. There the trajectories (varying frequency) of modal wavenumbers in the complex plane $\beta_z, \alpha_z$ using MA-DDA for Structures B-I, B-II, and B-III in Table 2 are given in the 23–30 THz frequency range. Similar conclusions to the ones drawn in regards to Structure A can be inferred by direct comparison of Figs. 4 and 7. One can see that there is a sudden change in the trajectories of Modes 1 and 2 passing from Structure B-II to B-III. It is apparent that in Structure B-IV after a certain frequency Mode 1 follows the original trajectory of Mode 2 and vice versa, i.e., they exchange roles, following each others’ trajectories for increasing frequency. This can be clearly observed by the close-up view of the first quadrant in Fig. 5. As it will be clear in the following, this mode degeneracy is due to both electric and magnetic scattering processes in all electromagnetic intersphere couplings [i.e., accounting for all the processes detailed in Eq. (7)], for large filling factors. Indeed, Fig. 6 shows the modes in Structure A-VI, which is the same plot in Fig. 4(f), without accounting for the electromagnetic coupling as described in Section 3.A; in other words, the dispersion diagram in Fig. 6 is obtained by using the MA-SDA-M procedure that is based on describing each microsphere only with its equivalent magnetic dipole, as was done in [11]. This means that the coupling term in Eq. (7) is neglected; hence, $\alpha_1 = 0$ is directly solved for complex wavenumber. The plot based on this approximation shows the absence of Mode 2. Therefore, mode degeneracy is not present in Fig. 6 as Mode 2 is not a solution when adopting the simple MA-SDA-M method and this clearly shows that the account of both electric and magnetic scattering processes in evaluating all electromagnetic intersphere couplings is essential for a proper description of the electromagnetic propagation in lattices as in Fig. 1.
permittivity difference between TiO$_2$ and PbTe (see Section 2). This also means that there is not a general rule for when mode degeneracy appears, and thus, such situations should be treated on a case-by-case basis.

The exchange of dominance here observed is enabled because the magnetic resonance is quite narrowband, i.e., it has a high quality factor, and the real part of $\alpha_{\text{mm}}$ changes sign and spans over a large range of values across the magnetic resonance (Fig. 2), changing the coupling scenario. In other words, for frequencies lower or higher than the magnetic resonance we may observe different signs in the scattering terms of the microspheres $\mathbf{G}^\infty_{\text{HM}} \cdot \mathbf{m}_0$ and $\mathbf{G}^\infty_{\text{EM}} \cdot \mathbf{m}_0$ since $\mathbf{m}_0 = \alpha_{\text{mm}} \mathbf{H}^\infty$, changing the interaction scenario when going across a magnetic resonance. Note that only terms with $\alpha_{\text{mm}}$ are modified, and the fields due to $\mathbf{G}^\infty_{\text{HM}} \cdot \mathbf{m}_0$ and $\mathbf{G}^\infty_{\text{EM}} \cdot \mathbf{m}_0$ change sign because $\mathbf{m}_0$ changes sign across the magnetic resonance if $\mathbf{H}^\infty_{\text{loc}} / \mathbf{r}_0 / \alpha_{\text{mm}}$ does not change sign. However, $\mathbf{H}^\infty_{\text{loc}}(\mathbf{r}_0)$ may (or may not) change sign because it is given by $\mathbf{H}^\infty_{\text{loc}}(\mathbf{r}_0) = \mathbf{G}^\infty_{\text{HP}} \cdot \mathbf{p}_0 + \mathbf{G}^\infty_{\text{HM}} \cdot \mathbf{m}_0$, and only the part $\mathbf{G}^\infty_{\text{HM}} \cdot \mathbf{m}_0$ may change sign if $\mathbf{m}_0$ changes sign. This last field composition, and its sign, may depend on packing. In deciding the sign of $\mathbf{H}^\infty_{\text{loc}}(\mathbf{r}_0)$, as a combination of the above terms, frequency may also play a role. Note that the frequency of the magnetic resonance for PbTe is much higher than the frequency of TiO$_2$, because of the different sizes, and this may affect how field constituents $\mathbf{G}^\infty_{\text{HM}} \cdot \mathbf{m}_0$ and $\mathbf{G}^\infty_{\text{HP}} \cdot \mathbf{p}_0$ superimpose.

5. DOMINANCE OF MODES THROUGH FIELD FITTING PROCEDURE

The aim of this section is to provide evidence of mode dominance interchange, relative to the modes observed in previous sections. We perform full-wave simulations based on the finite element method [using the commercial software High Frequency Structure Simulator (HFSS) by Ansys Inc.] on systems made of nine layers (in the $z$ directions; see Fig. 8 for a schematic) of arrayed TiO$_2$ microspheres as Structure A-II (for which no degeneracy was observed) and Structure A-V (for which degeneracy was observed) in Table 1, stacked in the direction of propagation of the normally incident plane wave illumination (the magnetic field is polarized along $y$).

We compare in Figs. 9(a) and 9(b) and Figs. 9(d) and 9(e) the modal wavenumbers obtained in Section 4 using MA-DDA to the ones retrieved via the field fitting procedure described in
[11]. The latter is performed here by assuming that the magnetic field (sampled at the center of each sphere) could be approximated by the superposition of two direct (“+”) and two reflected (“−”) waves, pertaining to two modes with transverse polarization and complex wavenumbers \( k_{z,A} \) and \( k_{z,B} \) as

\[
H_j(n) = A_+ e^{i k_{z,A}(n-\frac{1}{2})} + A_- e^{-i k_{z,A}(n-\frac{1}{2})} + B_+ e^{i k_{z,B}(n-\frac{1}{2})} + B_- e^{-i k_{z,B}(n-\frac{1}{2})},
\]

where \( n = 1, \ldots, 9 \) denote the sampling location along \( z \), and weights \( A_+, A_-, B_+, B_- \) and wavenumbers \( k_{z,A} \) and \( k_{z,B} \) are all complex valued unknowns. We refer to the wavenumber that has the strongest weight as the “dominant mode,” and indeed, when one is dominant the other has a much weaker weight (Fig. 9). Note that Modes 1 and 2 in Fig. 4 may exchange the dominant weight depending on the frequency. In our fitting, however, \( k_{z,A} \) and \( k_{z,B} \) will always describe the dominant and nondominant modes, respectively. The fitting method in Eq. (9) may lead to less accurate estimations for the nondominant mode weight and wavenumber than for the dominant mode depending on the frequency. 

The wavenumber result pertaining to the “Fit-D” curve is dominant because we have obtained \( |A_+ A_-^* + A_- A_+^*| > |B_+ B_-^* + B_- B_+^*| \) larger than \( |B_+ B_-^* + B_- B_+^*| \), as shown in Figs. 9(c) and 9(f) by the ratio \( |A_+| \) larger than about 10 for the analyzed frequencies, and an asterisk indicates a complex conjugate operation. We also report the (dominant) wavenumber retrieved using the Nicolson–Ross–Weir (NRW) algorithm from scattering parameters for finite thickness structures [34–38], computed here through full-wave simulations of four layers of TiO2 microspheres following the steps outlined in [3,23].

Referring to Figs. 9(a) and 9(b), we note that the wavenumber retrieved as “Fit-D” is in good agreement with both the MA-DDA result (Mode 1) and the NRW result. The wavenumber results pertaining to “Fit-ND-1” and “Fit-ND-2” curves are also in fair agreement with the MA-DDA result (Modes 2 and 3). Discrepancies may also be attributed to the presence of multipolar contributions not accounted for in our MA-DDA formulation. In Fig. 9 we do not observe any mode degeneracy for Structure A-II as predicted by MA-DDA. This is in agreement with the results reported in [24], where only one dominant mode was observed.

Referring to Figs. 9(d) and 9(e), we note that the wavenumber “Fit-D” is again in good agreement with both the MA-DDA result (Mode 1 and Mode 2, for different frequency ranges) and the NRW result. In this case, however, mode degeneracy is strikingly evident and the dominant mode Fit-D as well as the NRW retrieved results indicate the dominant mode is either Mode 1 or Mode 2 obtained with MA-DDA, depending on the frequency. The wavenumber result pertaining to “Fit-ND-1” and “Fit-ND-2” curves are also in fair agreement with the MA-DDA result. Discrepancies may also be attributed to the presence of multipolar contributions not accounted for in our MA-DDA formulation. In summary, in this case, we do observe mode degeneracy for Structure A-V as predicted by MA-DDA. This is an important result that should be considered in further developments when using metamaterials as in Fig. 1 and when using retrieval methods like NRW that may lead to nonaccurate results.

We finally note that for both structures, around the periodic lattice bandgap at about 300 THz, the two modes may have comparable weights, depending on the filling fraction, indicating that simple homogenization procedures based on a single-mode operation are not applicable in that particular frequency range. In that particular region a more elaborated homogenization procedure, accounting for spatial dispersion, as those provided in [22,24], could be applied. Away from the bandgap, however, homogenization procedures are again applicable because only one dominant mode may be found, as shown in Fig. 9.

6. METAMATERIAL HOMOGENIZATION IN TERMS OF EFFECTIVE REFRACTIVE INDEX

Following the discussion in previous sections, given that at any given frequency one mode only, with transverse polarization, is dominant and able to propagate inside the lattice, we treat here the metamaterial as a homogeneous material with effective refractive index. In Fig. 10 we show homogenization results from MA-DDA by retaining only the dominant modal

![Fig. 8. Metamaterial made of a stack (along z) of N layers of arrayed microspheres as in Fig. 1, illuminated by a plane wave with normal incidence. Fields are sampled at each layer.](image-url)
branches and from NRW, for the three structures in Table 1. Homogenization based on the NRW method is based on what has been discussed already in Section 5-C of [11], in Appendix A of [23], and in [25], and we refer the reader to those publications for more details. In [11], [24], and [25], it has been shown that these 3D lattices exhibit artificial magnetism and it was assumed that only one mode was dominant. Here we have shown that this property is maintained because different modes exchange dominance. Hence, homogenization based on the information of the wavenumber of the dominant mode in the structure is simply found by \( n_{\text{eff}} = \frac{k_{\text{dom}}}{k_0} \), as was done in [2, 3, 11, 23, 25], where \( k_{\text{dom}} \) represents the \( k_z \) wavenumber of the dominant mode, which is either Mode 1 or Mode 2 in Fig. 9, depending on the frequency range.

While MA-DDA is able to distinguish between modes, though here homogenization is based only on the dominant one, the use of the NRW method implicitly assumes that only one mode is propagating in the materials. The two methods are in good agreement for both Structures A-II and A-V. Although some disagreement is observed around the bandgap at

![Fig. 9](image_url)

Fig. 9. Dispersion diagrams of \( \beta_z, \alpha_z \) versus frequency and weight ratio \( |A/B| \) for dominant and nondominant modes versus frequency for (a)–(c) Structure A-II and (d)–(f) Structure A-V in Table 1. Solid curves are calculated with the MA-DDA method. The dotted curve (black) is calculated via the NRW retrieval method and it is pertaining only to the dominant portions of the modes, and it appears as an individual mode. Black circles (dominant, D), red stars and blue squares (nondominant, ND-1 and ND-2) are calculated by using two independent field fitting procedures with Eq. (9), based on two different initial wavenumber guesses.

![Fig. 10](image_url)

Fig. 10. Effective refractive index retrieved by retaining only the dominant modal branches when using MA-DDA and by NRW method, for the three structures in Table 1.
~300 THz, where simple homogenization procedures may not be applicable, this comparison is a further justification that one mode only should be considered dominant elsewhere because of the agreement with the NRW method. Fairly good agreement is observed also for Structure A–VI, although the close vicinity of neighboring spheres due to high packing makes us infer that dipolar approximation may fail, justifying the discrepancies between the two methods. Similar results are obtained also for Structure B in Table 2, not reported for brevity.

7. CONCLUSION

We have used the dual dipole approximation method developed in [25], based on considering both electric and magnetic dipole scattering from particles, to thoroughly investigate the effect of electromagnetic coupling and array packing on mode propagation in two distinct 3D lattices, made by either TiO₂ or PbTe microspheres that exhibit a strong magnetic polarizability in the millimeter wave or terahertz region, respectively. It has been shown that at these frequency ranges, the 3D lattices exhibit artificial magnetism, i.e., magnetic permeability different than unity. A more thorough discussion about why it is possible to provide a simple homogenization scheme (based on single-mode propagation in the lattice) is presented here. Losses are included and the investigation adds to wavenumbers with complex values. We observe that multiple modes are supported at various given frequencies, and that for low filling fractions only one mode with low attenuation constant is found to be dominant, whereas for large filling fractions we found two modes with transverse polarization dominant, but in different frequency ranges, when electromagnetic coupling (generated by both electric and magnetic dipoles) is included. Despite finding two distinct dominant modes we also find that at any given frequency only one of these two is dominant. This observation is also supported by mode fitting the field calculated via full-wave simulations. This mode degeneracy has been observed only when accounting for the electromagnetic coupling generated by both electric and magnetic dipoles, further stressing the importance to account for such coupling for a proper description of the electromagnetic properties of arrays, as in Fig. 1. In certain particular frequency regions where two modes are equally important, more elaborated homogenization techniques accounting for spatial dispersion as those in [22,24] should be adopted. Analogous findings may also occur in double negative artificial materials made of two kinds of spheres, as in [22], depending on array packing.

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