Magnetoelectric resonant metamaterial scatterers

Seršić, I.

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Chapter 4

Response of periodic arrays: experiment versus lattice sum dipole predictions

We implement the magnetoelectric polarizability tensor derived in the previous chapter as a building block for an electrodynamic model that describes the collective response of scatterers in periodic arrays. This model rigorously accounts for dipole coupling in arbitrary periodic lattices by generalizing Ewald lattice sums [1] to deal with the lattice response to both $E$ and $H$. We reinterpret the transmission measurements in Chapter 2 on split ring arrays with different lattice spacings and explain the resonance shifts and width in terms of the lattice response. We find excellent correspondence between the data and the new theory.

4.1 Introduction

Scattering experiments on metamaterials are frequently done using periodic planar arrays of magnetoelectric scatterers with sub-diffraction lattice spacings. The chain of reasoning from measurement to effective media parameters generally leads from measured intensity reflection and transmission coefficients that are used to validate brute force FDTD simulations, which in turn lead to parameter retrieval on basis of calculated amplitude reflection and transmission coefficients [2–8]. Very recently several groups reported non-trivial coupling phenomena between split rings, depending on their density, their local lattice coordination and their relative orientation [9–16]. Essentially, the physics is determined by dipole-dipole coupling between split rings. Since split rings have cross sections far in excess of the lattice unit cell in typical metamaterials the coupling is very strong. Also, since split ring polarizabilities are comparable to the unitary limit, their coupling is essentially
electrodynamic. In other words, coupling is not only via near field $d^{-3}$ interactions, but also strongly via radiation into the far field. Indeed, lattice transmission data reported in Chapter 2 show strong superradiant broadening effects at high densities.

In Chapter 3, we have derived how the polarizability tensor $\alpha$ can be approximated for a single SRR. In order to make successful predictions for measurements on arrays, apart from knowing the polarizability $\alpha$ of individual scatterers, one needs to understand the collective behavior of the lattice. Here, we expand our electrodynamic model of Chapter 3 to calculate reflection and transmission coefficients of magnetoelectric scatterers arranged in a periodic lattice. The collective response of lattices of scatterers with a scalar electric polarizability has previously been derived in [1]. Therefore, the aim of this chapter is not to derive a new theory, but to validate the electrodynamic picture of SRR response in arrays from the already existing theory, but with a tensorial $\alpha$ [17]. Technicalities of the theory can be found in [1, 18, 19].

4.2 Lattice sum theory

In this work we consider the response to plane wave illumination of a 2D periodic lattice of point scatterers, which is defined by a set of lattice vectors $\mathbf{R}_{mn} = m\mathbf{a}_1 + n\mathbf{a}_2$, or equivalently a set of reciprocal lattice vectors $\mathbf{g}_{mn} = m\mathbf{b}_1 + n\mathbf{b}_2$, where $m$ and $n$ are integers, and $\mathbf{a}_{1,2}$ and $\mathbf{b}_{1,2}$ are real space and reciprocal space basis vectors, respectively. The response of a particle at position $\mathbf{R}_{mn}$ is selfconsistently
4.2. Lattice sum theory

set by the incident field, plus the field of all other dipoles in the lattice according to

\[
\begin{pmatrix}
p_{mn} \\
m_{mn}
\end{pmatrix} = \alpha \left[ \begin{pmatrix}
E_{\text{in}}(R_{mn}) \\
H_{\text{in}}(R_{mn})
\end{pmatrix}
\right] + \sum_{m' \neq m, n' \neq n} G^0(R_{mn} - R_{m'n'}) \begin{pmatrix}
p_{m'n'} \\
m_{m'n'}
\end{pmatrix}.
\]  
(4.1)

For plane wave incidence with wave vector $k_{||}$, using translation invariance of the lattice, we can substitute a Bloch wave form \( (p_{mn}, m_{mn})^T = e^{ik_{||} \cdot R_{mn}} (p_{00}, m_{00})^T \) to obtain

\[
\begin{pmatrix}
p_{00} \\
m_{00}
\end{pmatrix} = [\alpha^{-1} - G^\neq (k_{||}, 0)]^{-1} \begin{pmatrix}
E_{\text{in}}(R_{00}) \\
H_{\text{in}}(R_{00})
\end{pmatrix}.
\]  
(4.2)

Here, \( G^\neq (k_{||}, 0) \) is a summation of the free space \( 6 \times 6 \) dyadic Green function \( G^0 \) over all positions on the 2D periodic real space lattice barring the origin

\[
G^\neq (k_{||}, r) = \sum_{m, n \neq 0} G^0(R_{mn} - r)e^{ik_{||} \cdot R_{mn}}.
\]  
(4.3)

We will refer to the summation without exclusion of \( m = n = 0 \) as \( G(k_{||}, r) \). The combination of Eq. (4.2) and (4.3) is the lattice sum formulation that has previously been reported in [1] for scalar Green function lattice sums where it was implemented using Ewald’s technique [18]. We are not aware of any reported implementation of lattice sums for the \( 6 \times 6 \) dyadic Green function \( G^0 \). The key difficulty sits in the fact that the sum in Eq. (4.3) is poorly convergent since \( G^0 \) decays only as \( 1/R \), whereas the number of terms with radius \( R < R_{\text{cutoff}} \) grows as \( R_{\text{cutoff}}^2 \). Ewald summation is the technique to deal with these difficulties. We refer to the excellent review by C. M. Linton [18] for an explanation of the technique for scalar Green functions. The dyadic case is obtained simply by pulling the derivatives in Eq. (3.3) and (3.4) that relate scalar and dyadic summand into the sum.

Once \( p \) and \( m \) are calculated via Ewald summation, we would like to find far field reflection and transmission. To find the reflected and transmitted waves, we note that for an observation point \( r \) in the far field, the Green function due to a source at \( r' \) can be written as

\[
G^0(r - r') = k^2 \frac{\exp(ik|r - r'|)}{|r - r'|} M
\]  
(4.4)

where \( M \) is a dimensionless matrix with elements of order unity that only depends on the direction, not the length of \( r - r' \). For a simple electric dipole, \( M \cdot p \)
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quantifies $\hat{p} - (\hat{p} \cdot \hat{r})\hat{r}$, where $\hat{r}$ is the unit vector along $(r - R_{m,n})$ the observation vector. Taking the scattered field intensity as the sum over all lattice points

$$
\begin{pmatrix}
E(r) \\
H(r)
\end{pmatrix}
= \sum_{n,m} k^2 \frac{\exp(ik|r - R_{nm}|)}{|r - R_{nm}|} e^{ik||R_{n,m}} M_{n,m} \begin{pmatrix} P_{00} \\ m_{00} \end{pmatrix}
$$

(4.5)

we make the usual far-field expansion assumption that the orientational factor $M$ does not vary with $n, m$ and we substitute

$$
\frac{\exp(ik|r - R_{m,n}|)}{|r - r'|} = \frac{i}{2\pi} \int dq \frac{\exp(iq \cdot (r|| - R_{m,n}) + k_z z)}{k_z}
$$

(4.6)

with $k_z = \sqrt{k^2 - |q|^2}$ and where integration is over parallel wave vector $q$. Using the completeness relation of the lattice,

$$
\sum_{m,n} e^{ik||R_{mn}} = \frac{2\pi^2}{A} \sum_{m,n} \delta(k|| - g_{mn}),
$$

(4.7)

where $A$ is the area of the unit cell, one might recast the summation to reciprocal space. As a consequence, one retrieves diffracted orders in the far field of the form

$$
\begin{pmatrix}
E(r) \\
H(r)
\end{pmatrix}
= \sum_{g,|k^g| \leq k} \begin{pmatrix} E^g \\ H^g \end{pmatrix} e^{ik^g \cdot r}
$$

(4.8)

where the diffracted wave vectors are $k^g = (k|| + g, \pm \sqrt{k^2 - |k|| + g|^2}) = k(\cos \phi \sin \theta, \sin \phi \sin \theta, \cos \theta)$. The far fields associated with each order are

$$
\begin{pmatrix}
E^g \\
H^g
\end{pmatrix}
= \frac{2\pi i k}{A \cos \theta} M(\theta, \phi) \begin{pmatrix} p_0 \\ m_0 \end{pmatrix}
$$

(4.9)

Where the orientation matrix $M(\theta, \phi)$ is the orientation dependent matrix from [20] (Chapter 15). Since these are only scattered fields, one still needs to add the incident field to obtain the zero-order transmitted beam. Dividing with the incident field, one obtains the transmission and reflection coefficients that are related via $t = 1 + r$.

The transmission coefficient for $x$-polarized incidence and detection, for instance, is then simply given by

$$
t_{xx} = 1 + \frac{2\pi i k}{A \cos \theta} (M(\theta, \phi)[\alpha^{-1} - g^z(k||, 0)]^{-1})_{xx}.
$$

(4.10)
4.3 Results

We have calculated transmission for many sets of parameters for $\alpha$, to test whether the lattice sum model can mimic the response measured for $200 \times 200 \times 30$ nm SRRs in square and rectangular arrays that we reported in Chapter 2. As an example, we show transmission for square lattices in Fig. 4.1, with the same lattice spacings used in Chapter 2. Fig. 4.1 shows two clear dips in the transmission spectra for all lattice spacings. The dip at $\lambda = 1.5 \mu m$ is the previously discussed $LC$ resonance, while the second dip at $\lambda = 900$ nm is a higher order resonance attributed to the plasmons excited along the bottom arm of the SRR [9]. Here, we have used $2 \pi c/\omega_0 = 1.6 \mu m$, $\gamma = 8.3 \times 10^{13}$ s$^{-1}$, $\alpha_E = 3.6 V$, $\alpha_H = 1.6 V$ and $\alpha_C = 2.1 V$ for the $LC$ resonance [21]. To mimic the higher order resonance, we have inserted $2 \pi c/\omega_1 = 0.96 \mu m$, $\alpha_E = 1 V$, $\alpha_H = 0.5 V$ and $\alpha_C = 0.4 V$, where $V = 0.0012 \mu m^3$. We assume the lattice to be embedded in a medium with refractive index $n = 1.23$, i.e., the average of the air and glass index on either side of the SRRs. Fig. 4.1 shows that the lattice sum model can indeed qualitatively reproduce all salient features also observed in the experimental data. These features include a blue shift of the $LC$ resonance with increasing density, as well as significant broadening. A shoulder appears on the red side of the $LC$ resonance for the highest density. Such resonance splitting would be expected on basis of the fact that the $LC$ resonance of split rings has two, not one eigenpolarizabilities. This edge is not evident in the data, and would require further study with a spectrometer that extends further into the IR. Finally, we note that the exact shape of the higher order resonance depends on the assumed surrounding refractive index: this resonance overlaps with a grating diffraction resonance into the glass, and not into the air. Fair account of this would require a model that can deal with the interface.

In order to determine which, if any, set of polarizability parameters best describe the data, we have performed transmission calculations for both rectangular and square lattices, for a large set of parameters $\alpha_E$, $\alpha_H$, and $\alpha_C$. In this scan of parameter space, we have kept the resonance frequency and damping constant fixed, $2 \pi c/\omega_0 = 1.6 \mu m$ and $\gamma = 8.3 \times 10^{13}$ s$^{-1}$. Since our scatterers are found at an air-glass interface, we assume the lattice to be in a homogeneous medium of index $n = 1.23$. We extract both the center frequencies and the linewidth of the transmission resonance, and define the set of parameters $\alpha_E \alpha_H$, and $\alpha_C$ that best matches our data, as those that best fit center frequency and linewidth simultaneously. Fig. 4.2 shows center frequency and linewidth as measured in Chapter 2, together with the dependence associated with the set of parameters that best fit the data. We find that the data cannot be fitted reasonably at all with parameters outside the range $0.5 V < \alpha_{E,H,C} < 3.9 V$. Only within this range do our calculations reasonably reproduce the extinction cross section $\sigma_{ext}$ reported by Husnik et al. [22].
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Figure 4.2: (a) Frequency of the magnetic resonance versus lattice spacing. The frequency blueshifts when decreasing $d_y$ whether $d_y = d_x$ (black squares) or not (light gray triangles, $d_x = 500$ nm), while it redshifts when decreasing $d_x$ (dark gray circles, $d_y = 500$ nm). (b) Linewidth of the magnetic resonance versus lattice spacing (color coding as in (a)). Dashed lines in (a) are electrostatic theory, while full lines in (a) and (b) are electrodynamic theory based on lattice sums.

The best fitting parameters are $\alpha_E = 3.6V$, $\alpha_H = 1.6V$, and $\alpha_C = 2.1V$ with estimated errors of $\Delta\alpha_E = \pm 0.8V$, $\Delta\alpha_H = \pm 0.5V$, and $\Delta\alpha_C = \pm 0.4V$. Parameter values in the order of particle volume $V$ indicate that split rings are strong scatterers on resonance. Fig. 4.2(a) shows the density dependence of the center frequency predicted by the lattice sum model together with the values extracted from experiment, and with the static model represented as dashed lines. The lattice sum calculation notably reproduces the strong resonance redshift for side-side coupled split rings, as well as the strong blueshift for both square arrays and the top-bottom coupled structures. A notable difference with the quasistatic model in Chapter 2 is the strong blueshift with increasing density for the square lattice that is observed also in the data, but not so clearly in the static model. A calculation of resonance frequency and FWHM with zero magnetoelectric cross coupling is shown in Fig. 4.3 (a) and (b) (dotted line). The lattice sum calculation now shows a very different behavior for square lattices, as the blueshift now disappears. The normal incidence linearly polarized transmission spectra hence point to a significant magnetoelectric cross coupling term $\alpha_C \gg 0$ which we will quantify directly in experiments reported in Chapter 5. Fig. 4.2 (b) shows the FWHM versus lattice spacing as measured and calculated with the lattice sum model. Since the Ohmic damping does not depend on the coupling in the electrostatic model, the FWHM broadening with decreasing lattice spacing can only be explained by the radiation damping in an electrodynamic picture, which the lattice sum model fully takes into account.
Figure 4.3: (a), (b), and (c) are the comparison between the data and lattice sum calculations of the center frequency, resonance linewidth and the effective extinction cross section per split ring derived from on resonance transmission with and without the magnetoelectric cross coupling term. The full lines are lattice sum calculations with $\alpha_E = 3.6V$, $\alpha_H = 1.6V$, $\alpha_C = 2.1V$, dotted lines with $\alpha_E = 3.6V$, $\alpha_H = 1.6V$ and without the cross coupling term. The black dashed line in (c) indicates the cross section of a single split ring (from [22]).

So far we have used only the resonance shift and the resonance width, to quantify the polarizability tensor. An interesting question is if the same parameters also satisfactorily explain other parameters measurable in the experiment. In Fig. 4.3 (c) we plot the effective extinction cross section per split ring as a function of lattice spacing with magnetoelectric coupling (full line) and without (dotted line). While the trend of a marked increase of effective cross section with reduced density is evident, the effective extinction cross section is generally underestimated. We have found no set of $\alpha$’s that quite fits all three quantities center frequency, width and $\sigma_{eff}$ simultaneously. The fact that no set of parameters can be found that simultaneously fit the center frequency, width, and cross section, is likely due to the asymmetric dielectric environment. The presence of an interface can significantly redistribute scattered light and can furthermore alter the local density of states (LDOS) of scatterers and thereby their extinction [23]. This is an interesting outlook with which we hope to further expand the lattice sum theory in the future.

4.4 Conclusion

We have shown how a point dipole lattice sum calculation can be implemented for point scatterers arranged in a 2D periodic array. We have extended the theory to apply to any magnetoelectric scatterer, and as such, explain the frequency shifts and superradiant broadening of the resonance peak with lattice spacing as seen in transmission measurements. We conclude that the polarizability of split rings is large compared to particle volume, quantifying the intuition that split rings are strong scatterers. Moreover our lattice sum calculation shows that the strong blueshift of the resonance for square lattices points at strong magnetoelectric cross coupling.
Indeed, it appears that driving with an electric field is more effective in setting up a large magnetic dipole moment, than direct magnetic driving. We pursue a more direct experimental method to quantify this observation in Chapter 5.

We envision that the model we have proposed in this chapter can be further improved and extended to deal with interesting questions. Firstly, the renormalization of the polarizability and the far field response due to the presence of an interface would be required to improve quantitative matching with data. Moreover, one can envision designing magnetoelectric gratings that have diffractive orders overlapping with the $LC$ resonance. Such gratings could have interesting chiral properties, due to the inherent optical activity that the magnetoelectric coupling in split rings entail. Also, we envision that the theory can be extended to deal with finite stacks of 2D lattices. Such an approach would allow one to build a fully electrodynamically coupled model system to examine if and how $\epsilon$ and $\mu$ emerge as a metamaterial grows from a surface to a bulk material.

Finally, we compare our theory with a recent model by Decker et al. [24] that explains resonance frequency shifts and linewidth broadening from data obtained by oblique incidence excitations of SRR arrays by accounting for long-range interaction effects between the split rings. These are described in a Lagrangian static model modified to account for a finite size of the lattice via a phase lag between adjacent split rings in an array [24, 25]. Both theories explain the response of an array for both far-field and near-field interactions, and account for a phase gradient over the array (Bloch waveform, Eq. (4.2)). However, it is not obvious if the model by Decker et al. satisfies the optical theorem. Violation of energy conservation can fundamentally only be avoided if radiative damping is chosen self consistently according to Eq. (3.20). Our theory rigorously satisfies the optical theorem by introducing the radiative damping term in the polarizability. Further radiative damping is contained in the rigorous electrodynamic lattice sums which explains the superradiant broadening of the resonance. In contrast, the theory of Decker et al. only takes into account radiation of electric dipole moments in the plane of the array, while magnetic dipole moments are neglected. This is an important point since in this thesis we show that magnetic dipole moments have significant polarizabilities comparable to the particle volume. A consistent model should equally account for radiation by electric and magnetic dipoles. For normal incidence experiments on nondiffractive samples, the far field radiated by magnetic dipoles cancels. However, our model can deal with any incidence angle, and any diffraction case.
[17] The implementation of the $6N \times 6N$ dyadic Ewald summation required for the lattice sum theory to describe the response of arrays of magnetoelectric scatterers has been done by A.F.K.

[21] Evidently, the polarizabilities $\alpha_{E,H,C}$ are complex quantities, since we are dealing with resonant scatterers that furthermore contain radiation damping. Quoted values for extracted parameters are for the absolute value on resonance throughout this chapter.


