Controlling electric, magnetic and magneto-electric dipole coupling in split ring clusters

A. F. Koenderink , I. Sersic, F. Bernal Arango and A. Kwadrin

Center for Nanophotonics, FOM Institute for Atomic and Molecular Physics (AMOLF)
Science Park 102-104, 1098XG-Amsterdam, The Netherlands
Fax: + 31 20 754 7290; email: f.koenderink@amolf.nl

Abstract

Especially in the near infrared and close to the visible wavelength regime, the physics of metamaterials and plasmonics are closely related. In both fields, one essentially deals with very strongly scattering subwavelength objects with cross sections close to the fundamental unitary limit. We seek to apply the language of plasmonics to also understand metamaterials. To this end we developed a fully electrodynamic theory that provides quantitative cross sections, resonance linewidths, and reflection and transmission coefficients for arbitrary clusters of magneto-electric point scatterers. Required input for this theory is a magnetoelectric polarizability that generalizes the purely electric polarizability from plasmonics. In this contribution we present experiments and microscopic calculations that quantify the polarizability tensor of many planar metamaterial scatterers. Surprisingly, we find that a strong cross polarizability is common to all metamaterial scatterers proposed so far. The cross polarizability implies that huge and robust optical activity is common to all metal scatterers that simultaneously have a magnetic and electric dipole response. We discuss possible uses for magnetoelectric antennas for single molecule microscopy, plasmon hybridization and grating diffraction.

1. Introduction

The field of metamaterials has spawned a plethora of metal scatterers for the near-infrared and visible part of the spectrum that are as strongly scattering as plasmonic objects, yet with a strong interaction also with the magnetic field of light H. In plasmonics, a scatterer is typically classified by (1) its local field enhancement capability, as would be sensed in single molecule fluorescence or SERS, and (2) the strength and width of its scattering resonance, as would be observed in, e.g., dark field microscopy. To first order these quantities are contained in the 'polarizability'. The strength of scattering as gauged by polarizability (units of volume) or cross section can be benchmarked against a universal 'unitary limit' that only depends on the host wavelength λ, and which states that for any dipolar scatterer the extinction cross section is at most $\sigma_{\mathrm{ext}} = 3/2\pi\lambda^2$ (equivalently $|\alpha| \leq 3/2(\lambda/2\pi)^3$) [1]. Recent experiments, for instance by Husnik et al. [2], by Liu et al. [3] and [4] show that metamaterial building blocks such as split rings should be viewed as scatterers with a resonance close to the unitary limit, very much like plasmonic particles. However, for a metamaterial object the polarizability should be a six-dimensional tensor

$$\begin{pmatrix} \mathbf{p} \\ \mathbf{m} \end{pmatrix} = \begin{pmatrix} \alpha_E & \alpha_{EH} \\ \alpha_{HE} & \alpha_H \end{pmatrix} \begin{pmatrix} \mathbf{E}_{\mathrm{incident}} \\ \mathbf{H}_{\mathrm{incident}} \end{pmatrix} \tag{1}$$

subject to well-known Onsager symmetry constraints to satisfy reciprocity [5]. This form brings out the fact that one expects not just an electric dipole $\mathbf{p}$, but also a magnetic response $\mathbf{m}$. The off diagonal terms furthermore encode that an electric field could induce a magnetic dipole, and vice versa. For quasistatic descriptions of looped wire antennas, wire omega particles, and helical structures, such 6x6

ISBN 978-952-67611-2-1

polarizability tensors are well known [5]. We showed in Ref. [6] that this picture can be extended to an electrodynamically consistent theory of scattering, provided the polarizability tensor is set to satisfy the Sipe-Kranendonk form of the optical theorem $\frac{1}{2i}\left[\boldsymbol{\alpha}-\boldsymbol{\alpha}^{*T}\right]=\frac{2}{3}k^3\boldsymbol{\alpha}^{*T}\boldsymbol{\alpha}$.

2. Optical activity

Given a microscopic model for the polarizability as input, our simple dipole model can predict many features such as cross sections, transmission spectra, superradiant damping with increasing lattice density, the chirality of stereometamaterials, and so forth, quantitatively. However, sofar these comparisons took the polarizability as a 'fit parameter' [6]. Very recently we set up experiments to determine the polarizability of NIR metamaterial scatterers. [6] A particularly striking prediction of the point scattering model is that a strong co-polarizability will manifest itself as a handed response in the single scatterer extinction cross section under oblique incidence, even for non-chiral building blocks. This is a phenomenon known as pseudochirality, which has a long standing history in the study of linear and nonlinear light scattering by oriented molecules [7, 8, 9]. That such optical activity without 2D or 3D chirality is *allowed* for metamaterial arrays once the illumination breaks symmetry is a fact that was already established by Plum et al. [10]. In this work we are not concerned with the *existence*, but with the *strength* of the phenomenon, as that is a direct measure for the polarizability tensor. We report on transmission measurements [11] of very dilute lattices of split rings resonant at 1.5 μm. Remarkably, for circularly polarized incident beams the transmission depends asymmetrically on incidence angle. For one handedness the extinction reaches a maximum at positive incidence angles around 50°, while the split rings are essentially transparent at large negative angles. As the handedness of the excitation is reversed, the behavior with incidence angle relative to the sample normal is mirrored. We find an extinction cross section of 0.13 $\mu\mathrm{m}^2$ at normal incidence, that doubles towards positive, and halves towards negative incidence angle. In order to extract the polarizability tensor we take into account that we measured on periodic lattices of split rings. We have extended the well known electrodynamic lattice sum calculations reviewed in [12] to deal with 6×6 dipole moments and interactions. Due to lattice interactions, the polarizability is renormalized so that

$$\begin{pmatrix}\mathbf{p}\\ \mathbf{m}\end{pmatrix}=\frac{1}{\boldsymbol{\alpha}^{-1}-\mathcal{G}(\mathbf{k}_{||},0)}\begin{pmatrix}\mathbf{E}_{\text{incident}}\\ \mathbf{H}_{\text{incident}}\end{pmatrix}\quad\text{with}\quad\mathcal{G}(\mathbf{k}_{||},\mathbf{r})=\sum_{\mathbf{R}\neq 0}\boldsymbol{G}(\mathbf{r},\mathbf{r}+\mathbf{R})e^{i\mathbf{k}_{||}\cdot\mathbf{R}}\tag{2}$$

where $\boldsymbol{G}$ is the free space 6×6 Green dyadic that quantifies the electric and magnetic fields radiated by electric and magnetic dipoles. This formulation sums all retarded interactions throughout the lattice using exponentially convergent Ewald summation techniques to find the lattice sum $\mathcal{G}$. Furthermore, we will present extensions of the method to arbitrarily thick, but finite, 2D stacks of lattices, to lattices close to dielectric interfaces, and to calculation of band structures and local density of states of lattices.

3. Magnetoelectric coupling

On basis of comparison of this model to experiment [11], we find $\alpha_{E,xx}=6.4V$, $\alpha_{H,zz}=0.9V$ and $\alpha_{EH,xz}=i2.1V$, in units of the physical split ring volume V. Three facts stand out: Firstly, the split rings have a very strong electric response to electric driving, competitive with plasmonic scatterers. Secondly, the direct magnetic response is sizeable, though 7 times weaker than the electric response. Thirdly, the cross-coupling is stronger than the direct magnetic polarizability. In other words, it is easier to set up a large magnetic dipole moment via electric driving than via magnetic driving. Physically, this magnetic moment upon electric driving arises from the fact that once the capacitive gap is charged upon driving by E, the discharge a quarter cycle later gives rise to a current, and hence a magnetic dipole. The strict ordering $\alpha_E\gg\alpha_{EH}\gg\alpha_H$ known for chiral molecules ($\alpha_{EH}\sim 10^{-3}\alpha_E$) is absent here, as all three polarizabilities are large, and within one order of magnitude of each other. One can view the polarizability tensor as a 'phase space' of possible response functions that one can seek to realize using meta-atoms. This space is constrained by reciprocity (Onsager), and energy conservation.

We have argued in Ref. [6] that if one considers planar scatterers (effectively 2×2 polarizability) with just a single resonant circuit equation of motion to generate $\boldsymbol{\alpha}$, that then only co-polarizabilities are allowed that satisfy $0 \leq |\alpha_{EH}| \leq \sqrt{\alpha_E \alpha_H}$. Remarkably, the extracted polarizability of split rings in our experiments is almost exactly *on* this limit of maximum cross coupling. We performed Surface Integral Equation calculations [11, 13] for a large family of single, flat, mirror symmetric particles reported by other workers to be a magnetic ingredient for metamaterials. By projecting scattered near fields on vector spherical harmonics we determine the induced dipole moments for various incident fields, thereby retrieving α. Remarkably we have found *all* particles we tested to be on the limit $|\alpha_{EH}| = \sqrt{\alpha_E \alpha_H}$ [11]. Also, the calculated extinction cross sections all displayed large single building block optical activity under oblique incidence. While efforts in our group are ongoing to determine if other parts of polarizability space, i.e., that either have $|\alpha_{EH}| < \sqrt{\alpha_E \alpha_H}$ or even $|\alpha_{EH}| > \sqrt{\alpha_E \alpha_H}$ can be accessed, our current conclusion is that maximum cross coupling is ubiquitous $|\alpha_{EH}| = \sqrt{\alpha_E \alpha_H}$. Our further experimental efforts to test this hypothesis pursue measurements of the angular distribution of light scattered by single split rings driven by handed fields [14], optical activity in grating diffraction, and the study of split rings interacting with their own mirror image when held close to a reflector.

4. Conclusion and outlook

The result that cross-polarizability is almost inevitably very strong, on the one hand is unfortunate for metamaterials, since it implies that bi-anisotropy is intrinsically difficult to avoid. One would require a strategy to cancel out intrinsic cross coupling, for instance by using larger unit cells with rotated copies of the same building block. This route could jeopardize the requirement that unit cells are much below the wavelength in size. On the other hand, robust optical activity has very large potential for many applications. We aim at optical antennas that control handed emission and absorption of photons.

References

[1] P. de Vries, D.V. van Coevorden, and A. Lagendijk Point scatterers for classical waves *Reviews of Modern Physics*, vol. 70, p. 447-466, 1998.
[2] M. Husnik, M. W. Klein, N. Feth, M. König, J. Niegemann, K. Busch, S. Linden and M. Wegener, Absolute extinction cross-section of individual magnetic split-ring resonators, *Nature Photonics*, vol.2, p. 614, 2008.
[3] N. Liu, H. Liu, S. N. Zhu and H. Giessen, Stereometamaterials, *Nature Photonics*, vol. 3, p. 157-162 (2009).
[4] I. Sersic, M. Frimmer, E. Verhagen and A. F. Koenderink, Electric and magnetic dipole coupling in near-infrared split ring metamaterial arrays *Physical Review Letters*, vol. 103, p. 213902;1-4, 2009.
[5] A.N. Serdyukov, I.V. Semchenko, S.A. Tretyakov, A. Sihvola, *Electromagnetics of bi-anisotropic materials: Theory and applications*, Amsterdam: Gordon and Breach Science Publishers, 2001.
[6] I. Sersic, C. Tuambilangana, T. Kampfrath, and A. F. Koenderink, Magneto-electric point scattering theory for metamaterial scatterers, *Physical Review B* vol. 83, p. 245102:1-12 2011.
[7] R. L. Dubs, S. N. Dixit, and V. McKoy, Circular Dichroism in Photoelectron Angular Distributions from Oriented Linear Molecules, *Physical Review Letters* vol. 54, 1249 (1985).
[8] C. Westphal, J. Bansmann, M. Getzlaff, and G. Schönhense, Circular dichroism in the angular distribution of photoelectrons from oriented CO molecules, *Physical Review Letters* vol. 63, p.151 - 154 1989.
[9] T. Verbiest, M. Kauranen, Y. van Rompaey, A. Persoons, Optical Activity of Anisotropic Achiral Surfaces, *Physical Review Letters* vol. 77, p. 1456-1459 (1996).
[10] E. Plum, *Chirality and Metamaterials*, PhD thesis, University of Southampton, United Kingdom, 2010.
[11] I. Sersic, M. A. van de Haar, F. Bernal Arango, A. F. Koenderink, Ubiquity of optical chirality in planar metamaterial scatterers, *Physical Review Letters* vol. 108, 223903:1-5 (2012).
[12] F. J. García de Abajo, Colloquium: Light scattering by particle and hole arrays, *Reviews of Modern Physics* vol. 79, p. 1267 - 1290, 2007.
[13] A. M. Kern and O. J. F. Martin, Surface integral formulation for 3D simulations of plasmonic and high permittivity nanostructures, *Journal of the Optical Society of America B* vol. 26, p. 732-740 (2009).
[14] I. Sersic, C. Tuambilangana and A. F. Koenderink, Fourier microscopy of single plasmonic scatterers *New Journal of Physics* vol. 13, p. 083019:1-13, 2011.