Exact Multipolar Decompositions with Applications in Nanophotonics

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The multipole decomposition of electromagnetic sources is an important tool for the study of light–matter interactions in general, and optical materials in particular. Here, a report is given on recent progress in the multipolar decomposition of electromagnetic sources. First, the exact and simple expressions for the multipolar moments of electric current density distributions are reviewed, and then, the results are extended to multipolar moments of magnetization current density distributions due to intrinsic spin. The consideration of both electric and magnetic sources allows to establish the conditions for sources of pure handedness. Scripts are provided that facilitate the computation of multipolar moments of arbitrary order. The work and the included examples of use are placed in the context of nanophotonics and metamaterials, and an outlook for applications in these and other fields is provided.

1. Introduction and Outline

The multipolar decomposition of a spatially confined electromagnetic source distribution is a basic tool in both classical and quantum electrodynamics. The resulting set of numbers are called the multipolar moments, and completely characterize both the radiation of electromagnetic fields by the source, and the coupling of external fields onto it. The multipolar decomposition is important in scientific areas dealing with the interaction between the electromagnetic field and material systems. Examples in physics range from nuclear and atomic physics, through nanophotonics and metamaterials, to the analysis of radiation by objects of astrophysical size. In chemistry, the dipolar and quadrupolar polarizabilities of a molecule determine most of its electromagnetic properties. In electrical engineering, the multipole decomposition is used to design and analyze the radiation from antennas.

In here, we report on recent progress in multipolar decompositions. New analytical expressions for the multipole moments of a localized electric current density distributions have been derived using an approach different from the traditional ones. The relative simplicity of the expressions allows a transparent connection to the widely used long-wavelength approximations, and a straightforward computation of any higher order correction. We provide a set of scripts that help in the practical computation of both exact and approximated moments of arbitrary multipolar order. The scripts are available online at https://github.com/Rasoul-Alaee/exact_multipoles. This Progress Report also contains extensions of the previous results. We derive the expressions of the multipoles caused by the magnetization current densities due to intrinsic spin, the proportionality factor between multipolar moments of the source and multipolar moments of the radiated fields, and the conditions that electromagnetic sources need to meet in order to become sources of pure handedness. These extensions complete the framework and make it usable for more general situations.

While the analysis and results in this Progress Report have general validity, we will put emphasis on and provide examples of use in nanophotonics and metamaterials, where multipolar decompositions are a crucial tool.[11–6] The increasing trend of studying optical materials based on its induced multipole moments may be explained by different reasons.

First, the discussion of multipolar moments induced by an external field in some spatially localized distribution of material became important in the slip-stream of the developments revolving around metamaterials.[7–10] The availability of technology to structure matter on length scales of only a few tens of nanometers brought about the need to understand its interaction with the electromagnetic field.[11,12] Just as our understanding of natural materials starts with the analysis of atoms or molecules, the understanding of metamaterials starts with the analysis of the meta-atoms, i.e., the repeated inclusions that constitute metamaterials.[13,14] The multipole moments are just the right tool for the task. The discussion of complicated structures is boiled down to just a few numbers, the multipole moments, that capture their electromagnetic response.[15–18] The multipole moments are also the direct link to the effective description of metamaterials, as many homogenization theories...
require as an input the electric/magnetic polarizabilities of the inclusions.\textsuperscript{[19-23]}

Second, with time passing, the community appreciated that individual meta-atoms already host a plethora of effects\textsuperscript{[24]} on their own. With the purpose of controlling and manipulating light on the nanoscale, an individual structure needs to be suitably shaped.\textsuperscript{[25,26]} Then, interference effects among the fields emitted by different multipolar moments induced in it strongly affect the radiation of light. This coined the term of an optical nanoantenna. The interference can occur in the spectral domain, where effects such as Fano resonances,\textsuperscript{[27,28]} electromagnetically induced transparency,\textsuperscript{[29]} or perfect absorption\textsuperscript{[30-34]} have been discussed. Alternatively, the interference can occur in the spatial domain, where effects such as a directional light emission are important.\textsuperscript{[35–38]} Also, the ability to control the spatial motion of individual scatterers due to an induced force, and the rotational motion due to an induced torque thanks to carefully tailored external fields has become important as well.\textsuperscript{[19–42]}

Third, with even more time passing, an appreciation has been established that our ability to control the interaction of light with structured materials cannot just be used to control light itself but also the interaction of the structure with other matter on the nanoscale: Atoms, molecules, or artificial atoms such as quantum dots.\textsuperscript{[43,44]} This completes the understanding of an optical nanoantenna. It can be used not only to localize light from the far-field to the nanoscale, but also, following the principle of reciprocity, to enhance the far-field emission of a localized emitter in close proximity to the nano-antenna.\textsuperscript{[45]} Around the same time, emitters that do not just have electric dipolar transitions but that also sustain magnetic dipolar or electric quadrupolar transitions were explored.\textsuperscript{[46–50]} The interaction of such multipolar emitters with multipolar optical nanoantennas has been a huge playground for scientific explorations.\textsuperscript{[51–54]}

Fourth, the analysis of multipole moments has become particularly important in the development of all-dielectric meta-atoms, optical nanoantennas, metasurfaces, and metamaterials.\textsuperscript{[55–61]} These all-dielectric materials are developed with the purpose to observe effects of the light–matter interaction like those observed using their metallic counterparts, but with the appealing benefit of being practically free of absorption.\textsuperscript{[62,63]} Supported by a mature silicon technology, all the means were at hand to conduct interesting science. At the heart of many of the research is the exploitation of the interplay of different multipolar moments.\textsuperscript{[64–66]} Examples are the Huygen’s elements, where backscattering is suppressed thanks to a balanced induction of electric and magnetic multipole moments,\textsuperscript{[67–70]} and, when set in an array, they allow to continuously control the phase of the transmitted field between 0 and $2\pi$.\textsuperscript{[71]}

In all these developments, the proper understanding of the induced multipole moments in the structure of interest is important. For example, to predict at which spectral position the electric and magnetic dipole moments balance each other for back reflection suppression,\textsuperscript{[72]} to predict whether the force acting on the particle is pulling or pushing,\textsuperscript{[73]} to quantitatively determine the directional coupling of light near surfaces,\textsuperscript{[74]} or to determine whether the metamaterial has a positive or a negative effective permeability.\textsuperscript{[75]}

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When considering all the nanophotonics and metamaterial research that is based on multipole moments, it is surprising to appreciate that the actual assignment of multipole moments is not completely clear. A first look into the literature might suggest that many different types of multipoles exist. For example, a distinction is made between Cartesian and spherical multipole moments,\cite{76,77} or multipole moments evaluated from the currents or the fields,\cite{78} or irreducible versus renormalized.\cite{79} Also novel kinds of multipole moments have been suggested, such as toroidal multipole moments, as an independent degree of freedom next to the electric and magnetic ones.\cite{80-83}

Therefore, in this context, one of the purposes of this Progress Report is to condense the understanding and to present in a concise way the multipole expansion. It links in an unambiguous manner the properties of the sources to those of the fields, allows in a transparent way to make small size approximations and reach expressions often used traditionally, but also raises the awareness that this is just an approximation that fails as soon as sources have a size not anymore much smaller than the wavelength but only smaller than the wavelength. The expressions to be discussed do not predict new or different quantities than previously existing expressions on an equal level of approximation. Nevertheless, the concise treatment is timely as the understanding has been sufficiently matured and the new exact expressions are arguably simpler than previous ones.\cite{84-85}

In Section 2, we provide an overview of the methodologies and results in recent work,\cite{86,87} and contrast them with the traditional treatment. We provide the proportionality factor between the multipolar moments of the source and the multipolar coefficients of the radiated field. We extend the results to magnetic spin current density distributions, and discuss multipolar decompositions in the helicity basis, providing the conditions that result in electromagnetic sources of pure handedness. In Section 3, we provide the expressions for cross-sections as a function of the multipolar moments: total scattering, forward scattering, backward scattering, absorption, and extinction. We also provide two examples of application in nanophotonics. Section 4 contains the outlook and Section 5 the conclusions.

2. Derivation of Multipolar Moments

2.1. Real (r)-Space Derivation

The derivation of the electric and magnetic multipolar coefficients of the fields radiated by a source is practically identical in the books by Jackson (ref. [84], Chap. 9.10), and Blatt and Weisskopf (ref. [85], Appendix B, §4), which are referential texts in electrodynamics and nuclear physics, respectively. The derivation uses two complementary elements. One is the localized electric current density distribution $\mathbf{J}(\mathbf{r}, t)$, whose harmonic decomposition leads to monochromatic functions $\mathbf{J}_n(\mathbf{r})$ with explicit space ($\mathbf{r}$) and implicit time [exp(−i\omega t)] dependences. The other element are monochromatic electromagnetic field solutions of the homogeneous wave equation in spherical coordinates, i.e., the electric and magnetic multipolar fields (see Equation (4)). The multipolar fields are used as the basis to decompose the radiation from the current density. The electric $a_{nm}$ and magnetic $b_{nm}$ coefficients in the decomposition are obtained as real space (r-space) volume integrals of functions of the monochromatic current density. The superscript $f$ in $a_{nm}$ or $b_{nm}$ denotes the fields. Omitting the magnetic current density, we find (ref. [84], Equation (9.165))

$$a_{nm} = \frac{i k}{\sqrt{j(j+1)}} \int d^3 r \, j_1(kr) \mathbf{Y}_{jm}(\hat{\mathbf{r}}) \mathbf{L} \cdot \mathbf{J}_n(\mathbf{r})$$

and (ref. [85], Appendix B, §4, Equations (4.10)–(4.11))

$$b_{nm} = \frac{-k^2}{\sqrt{j(j+1)}} \int d^3 r \, j_1(kr) \mathbf{Y}_{jm}(\hat{\mathbf{r}}) \mathbf{L} \cdot \mathbf{J}_n(\mathbf{r})$$

(1)

(2)

The caret and tildes in the left hand sides of Equations (1) and (2) indicate that different units and conventions have been used. We denote $k = \omega/c$, $r = |\mathbf{r}|$, and $\hat{\mathbf{r}} = \mathbf{r}/|\mathbf{r}|$ the unit r vector, or angular part of r, in the sense that it is equivalent to two direction angles. The $j_1 (\cdot)$ are spherical Bessel functions, $Y_{jm}(\hat{\mathbf{r}})$ are the spherical harmonics as defined in ref. [84], Equation (3.53), (·)† denotes complex transposition, and the three components of the vector $\mathbf{L}$ are the (differential) angular momentum operators for scalar functions (ref. [84], Equation (9.102)). The $\mathbf{X}_{jm}(\hat{\mathbf{r}})$ are the functions defined, e.g., in ref. [84], Equation (9.919). We note that $\mathbf{a} \mathbf{b}$ denotes the complex scalar product between the two three-vector $\mathbf{a}$ and $\mathbf{b}$.

Regarding the multipolar decomposition of the source currents, we can find an r-space derivation in Walecka’s book (ref. [88], Chap. 7). The current densities are transition current densities between different states of an atomic nucleus. The resulting integrals (ref. [88], Equation (7.25)) are

$$\mathbf{a}_{jm} = \frac{1}{k} \int d^3 r \left[ \mathbf{L} \cdot \mathbf{J}_n(\mathbf{r}) \right] \mathbf{Y}_{jm}(\hat{\mathbf{r}})$$

$$\mathbf{b}_{jm} = \int d^3 r \, j_1(kr) \mathbf{X}_{jm}(\hat{\mathbf{r}}) \mathbf{L} \cdot \mathbf{J}_n(\mathbf{r})$$

(3)

where we have again omitted the magnetic current density. The bars in the left hand sides of Equation (3) indicate that different units and conventions have been used compared to Equations (1) and (2).

For source distributions embedded in an achiral homogeneous and isotropic environment, each of the multipole moments of the current source couples to a single multipolar field. There is always a one-to-one proportionality relationship between the electric and magnetic multipolar moments of the current density [$a_{nm}$, $b_{nm}$], and the multipolar coefficients of the radiated field [$a'_{nm}$, $b'_{nm}$]. The exact proportionality coefficients depend on the conventions in the decompositions and on the system of units. From now on, in this Progress Report we will use S.I. units and decompose the radiated electric field into multipolar fields as

$$\mathbf{E}_r(\mathbf{r}) = \sum_{j=-\infty}^{\infty} \sum_{m=-j}^{j} \left[ a'_{jm} \mathbf{N}_{jm}(\mathbf{r}) + b'_{jm} \mathbf{M}_{jm}(\mathbf{r}) \right]$$

(4)
which meet in the spherical shell that denotes the real part, and the three components of are completely determined by the spatial Fourier components where are the outgoing spherical Hankel functions. The properties and of the current density \( \hat{J}_m(\mathbf{r}) \) will be discussed later.

The polarization vectors of \( \mathbf{X}_{pm}(\hat{p}) \) and \( \mathbf{Z}_{pm}(\hat{p}) \) are tangential to the surface of the shell, i.e., orthogonal (transverse) to the momentum vector \( \hat{p} \). This is achieved by decomposing these spherical shell is depicted in panel (b). The figure is reprinted from ref. [91] where it has been published under the Creative Commons Attribution 4.0 International License. b) \( p \)-space: The relevant part of the current density and their corresponding multipolar field components has been recently taken, resulting in a different expression of the current density and their corresponding multipolar field coefficients.

A different route for the derivation of the multipolar moments of the current density and their corresponding multipolar field coefficients has been recently taken, resulting in a different expression.

\[
|E_\omega(\mathbf{r}), H_\omega(\mathbf{r})| |\mathbf{r}| > R
\]

where \( \mathbf{M}_{pm}^m(\mathbf{r}) = h_m^1(\mathbf{r})\mathbf{X}_{pm}(\hat{r}) \) and \( \mathbf{N}_{pm}^m(\mathbf{r}) = (1/\mathbf{r})\nabla \times \mathbf{M}_{pm}^m(\mathbf{r}) \) are the magnetic and electric multipolar fields, respectively, and \( h_m^1() \) are the outgoing spherical Hankel functions. The properties of \( \mathbf{M}_{pm}^m(\mathbf{r}) \) and \( \mathbf{N}_{pm}^m(\mathbf{r}) \) will be discussed later.

\subsection{2.2. Momentum (\(p\))-Space Derivation}

A different route for the derivation of the multipolar moments of the current density and their corresponding multipolar field coefficients has been recently taken, resulting in a different kind of integrals from those in Equations (1)–(3). The difference starts by taking an extra step in the decomposition of \( \mathbf{J}(\mathbf{r}, t) \) which is assumed to be spatially confined and embedded in an infinite, isotropic, and homogeneous medium characterized by permittivity \( \epsilon \) and permeability \( \mu \). Besides the time harmonic decomposition, a spatial Fourier transform is also done to get the energy-momentum components \( \mathbf{J}_m(\hat{p}) \) of the current density

\[
\mathbf{J}(\mathbf{r}, t) = \int \frac{d\omega}{\sqrt{2\pi}} \exp(-i\omega t) \mathbf{J}_\omega(\mathbf{r})
\]

\[
= \int \frac{d\omega}{\sqrt{2\pi}} \exp(-i\omega t) \int \frac{d^3\mathbf{p}}{(2\pi)^3} \mathbf{J}_\omega(\mathbf{p}) \exp(i\mathbf{p} \cdot \mathbf{r})
\]

where \( \Re \{ \cdot \} \) denotes the real part, and the three components of the momentum vector \( \mathbf{p} \) are real numbers. The lower limit of the integral in \( d\omega \) excludes the static case \( \omega = 0 \). Then, the key step is the exploitation of a result by Devaney and Wolf [89] at each frequency \( \omega \), the transverse electromagnetic fields outside the source are solely determined by the part of \( \mathbf{J}_\omega(\mathbf{p}) \) in the spherical shell domain that satisfies \( |\mathbf{p}| = \omega \sqrt{\mu} = o/c \), where \( c \) is the speed of light in the surrounding medium. It was shown in ref. [89] that this part of the current completely determines the electromagnetic field generated by the source at any point \( \mathbf{r} \) such that \( |\mathbf{r}| > R \), where \( R \) is the radius of a sphere enclosing the sources. This result has been extended beyond spheres to confining volumes of any smooth enough shape (see ref. [90], Chap. 9). Figure 1a depicts a spatially confined monochromatic source distribution and the fields that it generates outside the source region. We note that, in particular, the outside region includes the near field region.

According to the above discussion one can continue by considering only the components of \( \mathbf{J}_m(\mathbf{p}) \) on the spherical shell of radius \( |\mathbf{p}| = o/c \), which we denote by \( \mathbf{J}_m(\hat{p}) \). The symbol \( \hat{p} \) represents the angular part of the momentum vector \( \mathbf{p} \), i.e., the solid angle in the spherical shell. In essence, we have simplified the problem by shedding the unnecessary components \( \mathbf{J}_m(\mathbf{p}) \neq o/c \), which are certainly contained in \( \mathbf{J}_m(\mathbf{r}) \) [and hence in \( \mathbf{J}(\mathbf{r}, t) \)]. It is worth mentioning that, in the \( d^3\mathbf{r} \) integrals of Equations (1)–(3), these noncontributing components are filtered out by the spherical Bessel functions, which act as a delta distribution in momentum space selecting the appropriate shell (see ref. [86], Section 3).

One then needs to extract the multipolar moments from \( \mathbf{J}_m(\hat{p}) \). This is achieved by decomposing \( \mathbf{J}_m(\hat{p}) \) into the three families of multipolar functions defined on a shell (ref. [91], B.3)

\[
\mathbf{X}_{pm}(\hat{p}) = \frac{1}{\sqrt{j(j+1)}} \mathbf{L} Y_{jm}(\hat{p})
\]

\[
\mathbf{Z}_{pm}(\hat{p}) = i\mathbf{p} \times \mathbf{X}_{pm}(\hat{p})
\]

\[
\mathbf{W}_{pm}(\hat{p}) = \hat{p} Y_{jm}(\hat{p})
\]
Table 1. Multipoles functions on the shell/multipole fields in r-space: Polarization character and eigenvalues of $J^m J_z$, and parity $\Pi$. The symbol $\perp$ means transverse polarization. The symbol $\parallel$ means longitudinal polarization (see Equation 1b). For any $j$ and $m$ are integers, and $m = -j \ldots j$. For $X_{jm}(\hat{p})/M_{jm}(r)$ and $Z_{jm}(\hat{p})/N_{jm}(r)$, $j > 0$. For $W_{jm}(\hat{p})/L_{jm}(r)$, $j \geq 0$. \[\begin{array}{cccc}
 j & J_z & \Pi & \text{Polarization} \\
 X_{jm}(\hat{p})/M_{jm}(r) & j(j+1) & m & \perp \sqrt{V \times 0} = 0 \\
 Z_{jm}(\hat{p})/N_{jm}(r) & j(j+1) & m & \parallel \sqrt{V \times 0} = 0 \\
 W_{jm}(\hat{p})/L_{jm}(r) & j(j+1) & m & \parallel \sqrt{V \times 0} = 0 \\
\end{array} \]

Together, they form an orthonormal basis for vectorial functions on the p-shell. We can write
\[\tilde{J}_m(\hat{p}) = \sum_{j,m} a_{j,m} Z_{jm}(\hat{p}) + b_{j,m} X_{jm}(\hat{p}) + c_{j,m} W_{jm}(\hat{p}) \] (7)

The $\{Z_{jm}(\hat{p}), X_{jm}(\hat{p}), W_{jm}(\hat{p})\}$ are eigenstates of the total angular momentum squared $J^2$; the angular momentum along the $\hat{z}$ axis $J_z$, and the parity operator. We highlight that these properties coincide with those of the multipole fields in Equation (4). Table 1 contains their eigenvalues. There is one important difference: There are three multipolar families for the sources but only two for the fields. The multipolar functions in momentum space $X_{jm}(\hat{p})$ and $Z_{jm}(\hat{p})$ are associated to the multipolar fields $M_{jm}(r)$ and $N_{jm}(r)$ in real space, as can be shown by Fourier transformation. The third kind of multipolar fields $L_{jm}(r) = (1/k) \hat{V} \{h_j^1(kr)Y_{jm}(\hat{r})\}$, linked to $W_{jm}(\hat{p})$, does not appear in Equation (4). The reason for this mismatch is found in the polarization degree of freedom. As depicted in Figure 1b, the polarization of $X_{jm}(\hat{p})$ and $Z_{jm}(\hat{p})$ is transverse (orthogonal) to $\hat{p}$, while the polarization of $W_{jm}(\hat{p})$ is longitudinal (parallel) to $\hat{p}$. In r-space, this distinction corresponds to the distinction between divergence free (transverse) and curl free (longitudinal) functions (see the last column in Table 1). This distinction is crucial because the longitudinal part of the current density does not produce electromagnetic fields outside the source region (ref. [92], §13.3, pp. 1875–1877). This can be seen as a cancellation due to the charge-current continuity equation (ref. [93], Appendix B). Therefore, while there are three kinds of degrees of freedom in the sources, only two of them couple to the electromagnetic field outside the source region.

Let us now go back to Equation (7). Expressions for the multipolar moments $\{a_{j,m}, b_{j,m}, c_{j,m}\}$ which only involve p-dependent functions can be easily obtained as on-shell projections of the different basis functions onto $\tilde{J}_m(\hat{p})$ (ref. [93], Equation (7)). In many practical situations, though, the current density is more readily available in the p-space. For example, in nanophotonics it is very common to use a numerical solver of Maxwell’s equations for obtaining the total electric field inside some nanostructure $E_{\text{in}}(\hat{r})$ upon a given illumination. The field scattered by the nanostructure can then be seen as the field radiated by the displacement current induced in the nanostructure by the illumination, which can be written as
\[J_{\text{ind}}(\hat{r}) = -i \omega E_{\text{in}}(\hat{r}) \Delta \epsilon(\hat{r}) \] (8)

where $\Delta \epsilon = \epsilon_3(\hat{r}) - \epsilon_\infty$ is the difference between the permittivity of the nanostructure and the permittivity of the surrounding medium. For this and other cases, it is convenient to have expressions of the multipolar moments that involve $J_m(\hat{r})$ instead of $\tilde{J}_m(\hat{p})$.

Straightforward steps (see ref. [86], Section 3) lead to
\[\sqrt{\frac{2\pi}{4\pi}} q_{jm}^m = \sum_{\Delta \omega} (-i)^j \int \hat{p} Q_{jm}(\hat{p}) Y_{jm}(\hat{p}) \int d^3 r J_m(\hat{r}) Y_{jm}(\hat{r}) j_j(kr) \] (9)
where $\Delta \omega = -\pi \ldots \pi$, $q_{jm}^m$ stands for any of the $\{a_{jm}^m, b_{jm}^m, c_{jm}^m\}$, and $Q_{jm}$ for the corresponding $\{Z_{jm}, X_{jm}, W_{jm}\}$. The electric current density $J_m(\hat{r})$ is expressed in the spherical basis
\[J_m(\hat{r}) = \begin{pmatrix}
 J_0^m(\hat{r}) + iJ_\perp^m(\hat{r}) \\
 \frac{1}{\sqrt{2}} \left[ J_0^m(\hat{r}) + iJ_\perp^m(\hat{r}) \right] \\
 \frac{1}{\sqrt{2}} \left[ J_0^m(\hat{r}) - iJ_\perp^m(\hat{r}) \right]
\end{pmatrix} \] (10)

It can be shown that, only $T$ terms contribute to the magnetic multipoles $b_{jm}^m$, while only $P$ terms contribute to the electric and longitudinal multipoles. Equation (9) is an exact expression for the $\{a_{jm}^m, b_{jm}^m, c_{jm}^m\}$ coefficients in terms of integrals in both p and r-spaces. Crucially, the momentum-space integrals do not depend on the current density and can hence be precomputed. We provide scripts that perform precisely this task: Given a multipolar order $j$, the scripts compute all the momentum-space integrals that are needed to compute the $q_{jm}^m$ assuming that the real space integrals in Equation (9) are available. We note the presence of the spherical Bessel functions $j_j(kr)$ in Equation (9). As discussed, they select the relevant momentum shell out of $J_m(\hat{r})$.

It is also possible to work on Equation (9) to produce expressions with a more familiar appearance. This has been done for the dipole (ref. [86], Equations (20)–(22)) and quadrupolar orders (ref. [87], Equations (41), (60), and (67) in the Supporting Information). Here are the expressions for the electric dipole moment
\[\begin{pmatrix}
 a_{10}^m \\
 a_{11}^m \\
 a_{1-1}^m
\end{pmatrix} = \frac{1}{\pi \sqrt{3}} \int d^3 r J_m(\hat{r}) j_j(kr) \]
\[\begin{pmatrix}
 b_{10}^m \\
 b_{11}^m \\
 b_{1-1}^m
\end{pmatrix} = -\frac{\sqrt{2}}{2\pi} \int d^3 r \frac{\hat{r}}{\hat{r} \times J_m(\hat{r})} \] (11)

and for the magnetic quadrupole moment
\[\begin{pmatrix}
 b_{20}^m \\
 b_{21}^m \\
 b_{2-1}^m
\end{pmatrix} = \frac{5i}{\pi \sqrt{10}} \int d^3 r \frac{\hat{r}}{\hat{r} \times j_j(kr)} \] (13)
where \( \hat{\mathbf{r}} = \frac{\mathbf{r}}{|\mathbf{r}|} \) is expressed in the spherical basis

\[
\hat{\mathbf{r}} = \begin{bmatrix}
\hat{r}_x \\
\hat{r}_y \\
\hat{r}_z
\end{bmatrix} = \frac{1}{\sqrt{2}} \begin{bmatrix}
x + iy \\
y \\
x - iy
\end{bmatrix}
\]

(14)

It is in principle possible to produce these kinds of expressions for any multipolar order (see the Supporting Information, including tables up to electric and magnetic octupole moments), but the complexity of such derivations suggests that the automatic use of Equation (9) by means of scripts like the ones we provide is a more practical option for the general case.

Since we are keeping an eye on the practical utilization of multipolar decompositions, a word about complexity is now in order. The differential operators acting on the current densities in the expressions of Equations (1)–(3) endow them with a degree of complexity which reduces their convenience as starting points for further work, either analytical or numerical. In this respect, Grahn et al.\cite{17} recently showed that the differential operators can be eliminated, reducing the complexity inside the integrals to pointwise projections of the current density with unit vectors, and multiplications by spherical Bessel functions, associated Legendre polynomials, and their derivatives with unit vectors, and multiplications by spherical Bessel functions (ref. [17], Equations (15)–(16)). In Equation (9) the complexity is reduced to multiplication with spherical Bessel functions and spherical harmonics, which contain associated Legendre polynomials, and in Equations (11)–(13) further reduced to algebraic operations involving the current density and position vectors, and multiplication by spherical Bessel functions.

2.3. Spherical versus Cartesian

For fixed \( j \) and \( m = -j \ldots j \), the electric \( \{ a_{jm}^e \} \) and magnetic \( \{ b_{jm}^m \} \) sets of multipole moments can be seen as the components of two spherical tensors that are irreducible under spatial rotations. That is, upon a rotation of the source, the new \( \{ a_{jm}^e \} \) and \( \{ b_{jm}^m \} \) are a linear combination of the old \( \{ a_{jm}^e \} \) and \( \{ b_{jm}^m \} \) without any contribution from different \( \tilde{j} \neq j \). Additionally, the linear combinations are the \( \tilde{j} \)-dependent rotation rules given by the Wigner matrices. The \( \{ a_{jm}^e \} \) and \( \{ b_{jm}^m \} \) are also irreducible with respect to the parity operation, and their transformation properties can be deduced from Table 1.

Any spherical tensor can be expressed in the Cartesian basis, and vice versa. For \( j = 1 \) the conversion can be seen in Equations (10) or (14). The conversions for \( j = 2 \) and \( j = 3 \) can be found in ref. [94], Table 2. There exist recursive methods to compute the conversions for arbitrary orders (see ref. [94] and the literature cited therein). The spherical or Cartesian tensors are different ways of representing the same physical information. While the appropriate choice of one basis over the other can simplify a given task, the underlying physical content does not change.

2.4. From Currents to Fields and Vice Versa

We now provide the relationship between the multipolar moments of the current density \( \{ a_{jm}^e, b_{jm}^m \} \) in Equation (7) and the field multipolar coefficients \( \{ \hat{a}_{jm}^e, \hat{b}_{jm}^m \} \) in Equation (4). It can be shown as, in S.I. units

\[
\hat{a}_{jm}^e = -i(2\pi)^{j+1} \frac{1}{j!} \frac{\partial}{\partial \mathbf{r}} \end{\mathbf{J}}_{e,m}(\mathbf{r})
\]

(15)

where \( Z = \sqrt{4\pi\epsilon_0} \) is the impedance of the embedding medium. Equation (15) is reached using the contents of ref. [89], and accounting for the different factors in their and our definitions of Fourier transforms, and the fact that they use Gaussian units. Incidentally, in Gaussian units the relationship is

\[
\hat{a}_{jm}^e = \frac{-i}{Z} \hat{a}_{jm}^e,
\]

(16)

For convenience, we also provide the conversion from our conventions to those in Jackson’s book (ref. [84], Chap. 9). Jackson’s field multipole coefficients \( \{ \hat{a}_{jm}^e, \hat{a}_{jm}^m \} \) in Equation (1) are related to \( \{ \hat{a}_{jm}^e, \hat{b}_{jm}^m \} \) and \( \{ a_{jm}^e, a_{jm}^m \} \) as follows

\[
\hat{a}_{jm}^e = \frac{-i(2\pi)^{j+1}}{Z} \frac{\partial}{\partial \mathbf{r}} \end{\mathbf{J}}_{e,m}(\mathbf{r})
\]

(17)

The conversions in Equation (17) can be deduced by considering the field decomposition used by Jackson in ref. [84], Equation (9.122), which is slightly different than Equation (4) and reads

\[
\begin{equation}
\mathbf{E}_e(\mathbf{r}) = Z \sum_{j=1}^{\infty} \sum_{m=-j}^{j-1} \left[ i \hat{a}_{jm}^e \mathbf{N}_{e,m}(\mathbf{r}) + i \hat{b}_{jm}^m \mathbf{M}_{e,m}(\mathbf{r}) \right]
\end{equation}
\]

(18)

and an extra \( 1/\sqrt{2\pi} \) factor. This factor comes from the fact that in ref. [84], Equation (9.1) the time-dependent complex current is: \( \mathbf{J}(\mathbf{r},t) = c \mathbf{J}_o(\mathbf{r}) \exp(-i\omega t) \). On the other hand, the first line of Equation (5) shows that, to obtain the same complex current, our \( \mathbf{J}_o(\mathbf{r}) \) should be, except for a delta function in frequency, equal to \( \mathbf{J}_o(\mathbf{r}) \sqrt{2\pi} \).

2.5. Long Wavelength Approximations

When the electromagnetic size of the current density distribution, denoted by \( R \), is much smaller than the wavelength of the field \( (kR \ll 1) \), approximate expressions for electric and magnetic multipolar moments may be used. They are easily obtained from the exact expressions of Section 2.2. For a general multipolar order \( j \), the spherical Bessel functions in Equation (9) can be approximated by their Taylor expansions truncated to any desired order \( (kr)^{2j+1} \). The resulting integrals can then be combined with the provided scripts. For the dipolar and multipolar cases the expressions in Equations (11)–(13) can be alternatively used (see refs. [86], Section 4 and [87]). To order \( (kr)^3 \), the electric dipole moment reads

\[
\begin{bmatrix}
a_{11}^e \\
a_{10}^e \\
a_{01}^e
\end{bmatrix} = \frac{1}{\pi \sqrt{3}} \int \mathbf{d}^2 \mathbf{r} \mathbf{J}_o(\mathbf{r})
\]

(19)
To order \((kr)^2\), it reads

\[
\begin{bmatrix}
    a_{11}^m \\
a_{10}^m \\
a_{01}^m
\end{bmatrix}
= -\frac{1}{\pi \sqrt{3}} \int \! d^3 r \, J_m(r)
- \frac{1}{\pi \sqrt{3}} k^3 \int \! d^3 r \, \frac{1}{10} \left[ [r \times J_m(r)] \cdot r - 2 r^2 J_m(r) \right]
\]

while, also to order \((kr)^2\), the magnetic dipole moment reads

\[
\begin{bmatrix}
    b_{11}^m \\
b_{10}^m \\
b_{01}^m
\end{bmatrix}
= -\frac{1}{2\pi \sqrt{3}} k \int \! d^3 r \, r \times J_m(r)
\]

The reader will readily recognize the expressions in Equations (19) and (21) to be proportional to the traditional ones (e.g., ref. [95], Chap. 9 and [84], Chap. 9). The proportionality factors arise from the fact that in the derivations of e.g., ref. [84], Chaps. 9.1–9.3, the long wavelength approximation is made first and the moments are defined later. As a result of this, Jackson's definitions of Cartesian multipole moments do not coincide with just changing \(\{a^m_{ij}, b^m_{ij}\}\) from the spherical to the Cartesian basis. There are frequency and \(j\) dependent factors between the two sets. These factors, together with the change from spherical to Cartesian, are provided in the appendices of ref. [87] for both dipolar and quadrupolar cases. We note that the Cartesian multipole moments in ref. [87] are defined in a way that can be directly compared with the definitions in ref. [84], Chaps. 9.1–9.3.

We now focus on the second term in Equation (20). It is often called toroidal dipole.

2.6. Toroidal Multipoles

The toroidal multipoles were first introduced in the static case.[96] Later, their dynamic counterparts were presented as a new independent multipole family, besides the electric and magnetic ones.[97,98] The interest on dynamic toroidal multipoles due to intrinsic spin has recently experienced a renewed growth in the nanophotonics and metamaterials community.[78,82,99–103] The original claim that the toroidal multipoles are a third independent family of multipoles has been picked up in some of the recent work. Some consequent predictions have been made, like for instance the existence of spectroscopic resonances of toroidal character, in addition to electric and magnetic resonances. The exact expressions (see ref. [86], Equation (21) and Table II in ref. [87]) and their approximations (ref. [86], Equation (28)) and Table I in ref. [87]) allow to critically analyze the original claim and show that is wrong.[93] The bottom line is that the toroidal multipoles are higher order corrections in the long wavelength approximation of the exact electric multipoles, and that there is no independent toroidal degree of freedom in electromagnetism. This, in particular, means that there are no spectroscopic resonances of a new third kind.

2.7. Multipole Moments due to Intrinsic Spin

There is a different kind of electromagnetic source besides electric current densities: Magnetic current densities due to the intrinsic spin of, for example, nucleons or electrons. Intrinsic magnetization current densities of protons and neutrons are often considered in atomic and nuclear physics (see, e.g., ref. [88], Equation (7.26)). The electron spin gives rise to the intrinsic magnetic response of some materials to electromagnetic fields at GHz frequencies and below.[104,105] We have purposely left the intrinsic magnetization current densities out of the discussion because the multipole moments that they originate can be computed by an appropriate use of the expressions for the multipole moments originated by the electric current densities. This can be shown in a heuristic way by examining existing expressions for the total multipole moments of the sources containing both electric and magnetic current densities, like ref. [88], Equation (7.25) which can be written as

\[
\begin{align*}
\bar{a}^m_{\text{trans}} &= \int \! d^3 r \left[ \frac{1}{(kr)^3} \right] \cdot \left[ J_m(r) + k^2 \left[ j \cdot (kr) X_m(\hat{r}) \right] \right] M_m(r), \\
\bar{b}^m_{\text{trans}} &= \int \! d^3 r \left[ j \cdot (kr) X_m(\hat{r}) \right] J_m(r) + \left[ V \times j \cdot (kr) X_m(\hat{r}) \right] M_m(r)
\end{align*}
\]

where \(M_m(r)\) is the spin current density, which is defined in the same way as in ref. [84]. Let us write more concise expressions for the multipole moments in Equation (22) as a function of the two kinds of sources

\[
\begin{align*}
\bar{a}^m_{\text{trans}} \left[ J_m(r), M_m(r) \right], \quad \bar{b}^m_{\text{trans}} \left[ J_m(r), M_m(r) \right]
\end{align*}
\]

where \(J_m(r)\) is the transverse part of the current, that is, without the contributions of the longitudinal degrees of freedom. We can use \(J_m(r)\) instead of \(J_m(r)\) because Equation (22) projects \(J_m(r)\) onto transverse fields only, as do Equations (1)–(3). It is easy to see that

\[
\begin{align*}
\bar{a}^m_{\text{trans}} [0, M_m(r)] &= \bar{b}^m_{\text{trans}} [k M_m(r), 0] \\
\bar{b}^m_{\text{trans}} [0, M_m(r)] &= \bar{a}^m_{\text{trans}} [k M_m(r), 0]
\end{align*}
\]

The same relationship holds for the multipole moments in our conventions \(\{a^m_{ij}, b^m_{ij}\}\). This allows to compute the electric and magnetic multipolar moments due to \(M_m(r)\) by using the expressions for the moments due to \(J_m(r)\).

The use of the helicity operator \((\hat{p} \times \hat{p})\) in p-space, or \((1/k \hat{V} \times \hat{r})\) in r-space) provides another viewpoint onto Equation (24). The helicity operator is the generator of the electromagnetic duality transformation, which exchanges electric and magnetic quantities.[106,107] When applied to Equation (7) it results in

\[
\hat{p} \times J_m(\hat{p}) = \sum_{m} b^m_{\text{trans}} Z_m(\hat{p}) + a^m_{\text{trans}} X_m(\hat{p})
\]

where one uses that \(\hat{p} \times Z_m(\hat{p}) = X_m(\hat{p})\), \(\hat{p} \times X_m(\hat{p}) = Z_m(\hat{p})\), and \(\hat{p} \times W_m(\hat{p}) = 0\). The exchange of electric and magnetic quantities is obvious in Equation (25). It can be checked that the factor of \(k\) multiplying \(M_m(\hat{r})\) in Equation (24) makes the (S.I.) units of \([\hat{p} \hat{p} J_m(\hat{p})]\) match those of a magnetic current density in the shell. We note that the disappearance of the longitudinal terms does not pose a problem. \(M_m(\hat{r})\) does not have
longitudinal components because the divergence of $\mathbf{M}_o(r)$ is zero since there are no magnetic charge monopoles, but rather intrinsic magnetic dipoles.

### 2.8. Sources of Pure Handedness

The change of basis

$$
\begin{align*}
    g_{jm}^0 &= \frac{a_{jm}^+ + b_{jm}^-}{\sqrt{2}} \\
    g_{jm}^+ &= \frac{a_{jm}^+ - b_{jm}^-}{\sqrt{2}} \\
    g_{jm}^- &= \frac{a_{jm}^- - b_{jm}^-}{\sqrt{2}}
\end{align*}
$$

transforms objects of well-defined parity (e.g., electric and magnetic), into objects of well-defined helicity, i.e., generalized polarization handedness. For the choice of the signs in the helicity multipoles, we follow ref. [108], Equations 11.4–(6), 11.4–26 (27). In this way

$$
\begin{align*}
    A_{a0jm}(r) &= \frac{1}{\sqrt{2}} \left[ N_{jm}(r) + M_{jm}^+(r) \right] \\
    A_{a0jm}(r) &= \frac{1}{\sqrt{2}} \left[ N_{jm}(r) - M_{jm}^+(r) \right]
\end{align*}
$$

are outgoing spherical waves of pure handedness, meaning that their angular spectrum contains only plane waves (either propagating or evanescent) of a single polarization handedness: Left handed for helicity +1 and right handed for helicity −1. In the momentum shell representation of the current densities, the combinations

$$
\begin{align*}
    K_{jm}^+(\hat{p}) &= \frac{1}{\sqrt{2}} \left[ Z_{jm}(\hat{p}) + X_{jm}(\hat{p}) \right] \\
    K_{jm}^-(\hat{p}) &= \frac{1}{\sqrt{2}} \left[ Z_{jm}(\hat{p}) - X_{jm}(\hat{p}) \right]
\end{align*}
$$

are the sources of pure handed radiation, whose near fields in particular also contain a single handedness. By reciprocity, they are also responsible for the coupling to each of the two helicities of the electromagnetic field.

When a monochromatic electromagnetic field contains a single helicity, only one of the two Riemann–Silberstein combinations

$$
\sqrt{2} G_{jm}^0(\mathbf{r}) = E_{a0}(\mathbf{r}) + iZH_{a0}(\mathbf{r})
$$

is different than zero. The other one is zero for all $\mathbf{r}$ (see ref. [111], Figure 1). This kind of fields are desirable for the near-field enhanced differential interaction with chiral objects, e.g., chiral molecules. It should be noted that the commonly used expression for the local “chirality density” of the field

$$
C_a(\mathbf{r}) = -\frac{\varepsilon_0}{2} \text{Im}\{E_{a}(\mathbf{r}) \cdot B_{a}(\mathbf{r})\}
$$

where Im\{·\} denotes the imaginary part, can also be written in the arguably more suggestive form

$$
C_a(\mathbf{r}) = \frac{\varepsilon_0}{4c^2} \left( |G_{a}(\mathbf{r})|^2 - |G_{a}^*(\mathbf{r})|^2 \right)
$$

We will finish this section by determining the conditions that $\mathbf{J}_o(r)$ and $\mathbf{M}_o(r)$ must meet in order to produce a source of pure helicity.

If a confined electromagnetic source with both $\mathbf{J}_o(r)$ and $\mathbf{M}_o(r)$ is to produce pure handed ($±1$) radiation, Equation (26) indicates that either of two conditions must be met

$$
a_{jm}^+ = b_{jm}^+ \text{ for all } (j,m), \text{ or } a_{jm}^- = -b_{jm}^- \text{ for all } (j,m)
$$

Using the notation of Equation (23) we write then

$$
a_{jm}^+ \mathbf{J}_o(r) = \pm b_{jm}^+ \mathbf{M}_o(r)
$$

One can use Equations (23)–(24) and the linearity and homogeneity of the $a_{jm}^+[-]$ and $b_{jm}^-[-]$ functions in each of their arguments to show[115] that Equation (33) can be written as

$$
a_{jm}^+ \mathbf{J}_o(r) = \pm k \mathbf{M}_o(r)
$$

There are different ways to meet Equation (34). The most obvious one is

$$
\mathbf{J}_o(r) = \mp k \mathbf{M}_o(r)
$$

We note that that both kinds of sources, electric and magnetic, must be present to meet Equation (35). The general way to find all the possibilities for pure handedness is by deducing[116] from Equation (25) that

$$
a_{jm}^+ \mathbf{F}_o(r) = \pm b_{jm}^+ \left[ \frac{\nabla \times \mathbf{E}_o(r)}{k} \right]
$$

whose first line, applied to Equation (34), tells us that the electromagnetic source will be of pure ±1 helicity when

$$
\frac{\nabla \times \left[ \mathbf{J}_o(r) \mp k \mathbf{M}_o(r) \right]}{k} = \pm \left[ \mathbf{J}_o(r) \mp k \mathbf{M}_o(r) \right]
$$

Therefore, besides the solutions in Equation (35), the source will have a definite positive handedness if

$$
\frac{\nabla \times \mathbf{J}_o(r)}{k} = \mathbf{J}_o(r) \text{ and } \frac{\nabla \times \mathbf{M}_o(r)}{k} = \mathbf{M}_o(r), \text{ or }
$$

$$
\frac{\nabla \times \mathbf{J}_o(r)}{k} = -k \mathbf{M}_o(r), \text{ which implies } \frac{\nabla \times k \mathbf{M}_o(r)}{k} = -\mathbf{J}_o(r)
$$

and will have a definite negative handedness if

$$
\frac{\nabla \times \mathbf{J}_o(r)}{k} = -\mathbf{J}_o(r) \text{ and } \frac{\nabla \times \mathbf{M}_o(r)}{k} = -\mathbf{M}_o(r), \text{ or }
$$

$$
\frac{\nabla \times \mathbf{J}_o(r)}{k} = k \mathbf{M}_o(r), \text{ which implies } \frac{\nabla \times k \mathbf{M}_o(r)}{k} = -\mathbf{J}_o(r)
$$
The implications in the second lines of Equations (38) and (39) follow because, for transverse fields, \( \left( \frac{\nabla \times}{k} \right) \) is the identity operator. These lines are identical. The solution to this apparent contradiction is that the multipoles due to the electric current density cancel exactly the multipoles of the magnetic current density. This possibility exists because, according to Equation (32), we are looking for sources of a pure \( \pm 1 \) handedness by nulling the sources of the opposite one. The second lines of Equations (38) and (39) null both of them.

In summary, sources of pure \( \pm 1 \) handedness are obtained when

\[
\mathbf{J}_r^\omega (\mathbf{r}) = \pm i k \mathbf{M}_r(\mathbf{r}), \quad \text{or} \quad \frac{\nabla \times}{k} \mathbf{J}_r^\omega (\mathbf{r}) = \pm \mathbf{J}_r^\omega (\mathbf{r}) \quad \text{and} \quad \frac{\nabla \times}{k} \mathbf{M}_r(\mathbf{r}) = \pm \mathbf{M}_r(\mathbf{r})
\]

We observe that sources of a single kind, either electric or magnetic, can be of pure handedness by meeting the second line of Equation (40), with the other kind of source being equal to zero. This is what happens, approximately, for the \( \mathbf{J}_r^\omega (\mathbf{r}) \) induced in quasi-dual dielectric objects\(^{70,117} \) by a field of pure helicity (i.e., either \( \mathbf{G}_r^\omega (\mathbf{r}) = 0 \) or \( \mathbf{G}_{-r}^\omega (\mathbf{r}) = 0 \)). Note that chirality of the object is not necessary for it to host an induced source of pure handedness. The effect is caused by the chirality of the illuminating field and the fact that a dual object does not couple the two helicities of the field,\(^{116,111} \) which means that an incoming field of a given helicity cannot induce a source of the opposite one.

### 3. Application in Nanophotonics

As previously mentioned, the following situation is very common in current nanophotonics research. Given an object and an incident illumination, often a plane wave, a numerical tool is used to solve Maxwell’s equations. In particular, the field inside the object becomes available. This internal field can be seen as an induced current distribution as per Equation (8), which then reradiates the scattered field. The scattered field is decomposed as in Equation (4). The incident electric field can be expressed as a linear combination of multipolar fields similar to Equation (4)

\[
\mathbf{E}_{\text{inc}}^\omega (\mathbf{r}) = \sum_{j=1}^{\infty} \sum_{m=-j}^{j} \left[ f_p^{\omega m} \mathbf{N}_m^\omega (\mathbf{r}) + f_q^{\omega m} \mathbf{M}_m^\omega (\mathbf{r}) \right]
\]

where here the multipolar fields contain spherical Bessel functions instead of spherical Hankel functions.

Then, the scattered, extinction, and absorption cross-sections can be computed as

\[
\sigma_{ns} = \frac{1}{k} \sum_{j=0}^{\infty} \sum_{m=-j}^{j} \left| f_p^{\omega m} \right|^2 + \left| f_q^{\omega m} \right|^2
\]

\[
\sigma_{ext} = \frac{1}{k} \sum_{j=0}^{\infty} \sum_{m=-j}^{j} \mathbb{R} \left[ \left( f_p^{\omega m} \right)^* \left( f_q^{\omega m} \right) + \left( f_q^{\omega m} \right)^* \left( f_p^{\omega m} \right) \right]
\]

\[
\sigma_{abs} = \sigma_{ext} - \sigma_{ns}
\]

The minus sign in \( \sigma_{ns} \) appears because neither of our definitions of incident field [Equation (41)] or scattered field [corresponding to Equation (4)] contain a minus sign. We have also chosen to omit an overall scaling factor \( |E_0| \) that is often included in the multipole expansions of the incident and scattered fields. These factors cancel when computing cross-sections since the reradiated powers are divided by the incident intensities.

The forward and backward scattering cross-sections are proportional to the normalized intensity that is radiated onto the forward and backward directions by a given scattered field represented by its \{ \( f_p^{\omega m}, f_q^{\omega m} \) \} coefficients, as in Equation (4). The cross-sections can be computed by summing the norm squared of the projections of the scattered field onto the multipolar representation of RCP and LCP plane waves. The electric \( \{ f_p^{\omega m} \} \) and magnetic \( \{ f_q^{\omega m} \} \) multipolar expansion coefficients for circularly polarized plane waves with momentum along the \( \hat{z} \) direction, which we take here as forward direction read

LCP: \( f_p^{\omega m} = f_{1m} = i \delta_{1m} \sqrt{2} \pi (2j+1) \)

RCP: \( f_q^{\omega m} = -f_{1m} = -i \delta_{1m} \sqrt{2} \pi (2j+1) \)

where \( \delta_{1m} \) is the Kronecker delta, and along the \(-\hat{z} \) direction (backward)

LCP: \( f_p^{\omega m} = f_{1m} = (-i) \delta_{1m} \sqrt{2} \pi (2j+1) \)

RCP: \( f_q^{\omega m} = -f_{1m} = (-i) \delta_{1m} \sqrt{2} \pi (2j+1) \)

The forward scattering cross-section then reads

\[
k^2 \sigma_{\text{fs}}^{\omega m} = \sum_{j=0}^{\infty} \sqrt{2} \pi (2j+1) (-i) \left( f_p^{\omega m} + f_q^{\omega m} \right)^2
\]

and the backward scattering cross-section

\[
k^2 \sigma_{\text{bs}}^{\omega m} = -\sum_{j=0}^{\infty} \sqrt{2} \pi (2j+1) (i) \left( f_p^{\omega m} + f_q^{\omega m} \right)^2
\]

Expressions for the induced force and torque can be found in the Supporting Information of ref. [41], where the \{ \( f_p^{\omega m}, f_q^{\omega m} \) \} are minus the ones defined here.

### 3.1. Examples

We first use Mie theory to verify the exact multipole moments, i.e., Equations (11)–(13) (or Table II in ref. [87]). The Mie theory allows to compute the scattering cross-sections of a sphere by using the individual contributions of each induced electric and magnetic multipole moment when illuminated by a plane wave. Note that the scattering cross-sections computed by the Mie theory are without any approximation and are valid for any wavelength and size of the sphere.
Let us consider a high-index dielectric sphere in free space. The sphere is illuminated with a linearly $x$-polarized plane wave that propagates in the $z$-direction. The induced multipole moments are computed once by using the Mie theory, and also using Equations (11)–(13) (or Table II in ref. [87]). In order to compute the electric field distribution inside the sphere, we use a numerical finite element solver (COMSOL).

Figure 2a shows the contributions of different multipole moments to the scattering cross section as a function of the particle’s size parameter $2a/\lambda$. The permittivity of the dielectric sphere is assumed to be: thickness $t=40$ nm, height $h=350$ nm, and radius $a=250$ nm. For example, for a dipolar particle (i.e., $j=1$), the maximum cross section is $3\lambda^2/2\pi$.[119]

Figure 2b shows the total scattering and absorption cross sections of the dielectric sphere. Note that the contribution of each multipole moment to the scattering cross section is normalized to $\lambda^2/2\pi$. For a spherical particle, there is a universal limit for each multipole, i.e., $(2j+1)\lambda^2/2\pi$. For example, for a dipolar particle (i.e., $j=1$), the maximum cross section is $3\lambda^2/2\pi$.[119]

and cannot be used for large particles (compared to the wavelength). This large deviation in multipole moments obtained from long-wavelength approximation will lead to incorrect results in any physical quantities obtained from the these multipole moments. The exact expressions have been recently used to study scattering properties and optical forces exerted on various particles.[76,120–124]

As the last example, we analyze a chiral structure under circularly polarized illumination. Chiral structures and chiral metamaterials are a subject of intense research, mostly because their ability to differently affect the two helicities of the field. For such kind of analysis, the helicity multipole formalism is specially well suited. Indeed, Figure 4 shows that the considered chiral cross-wire interacts more with light of positive helicity.

3.2. Discussion of Selected Applications

We emphasize at this point that any physical quantity (e.g., scattering, absorption, or extinction cross sections, optical forces and torques and many others) obtained from the exact multipole moments will be exact as well. The ability to precisely determine the multipole moments is therefore decisive in a larger number of current research strands in the field of nanophotonics where these expressions are applied. As a referential example we may mention here the field of optical nanoantennas that has been shortly discussed already in the introduction.[1]

Optical nanoantennas offer excellent control over the radiation characteristics if a proper understanding of the induced multipole moments is exploited.[33,126,127] For example, quantum emitters that are placed close to an optical nanoantenna can be forced to radiate in a specific direction.[35,45]
Another application can be found in computational electromagnetism. In some computational techniques like the discrete dipole approximation, the induced multipole moments allow to observe various phenomena such as a suppressed backscattering,[26,31,72,128] Fano resonances,[27,28] perfect absorption,[30,32,34] or a plasmonic analogue to electromagnetically induced transparency.[29] All these aspects are particularly important in the emerging field of all-dielectric photonic nanostructures, as it has been also mentioned already in the introduction. In this field of research it has been appreciated that higher order multipole moments can be induced with a notable amplitude in suitably designed scatterers made from a high permittivity material such as silicon. Shaping the dispersion of these multipole moments is the key to control light scattering. In particular, for high index scatterers the multipole moment that is driven at the longest wavelength into resonance is not the electric dipole but the magnetic dipole moment.[129] This allows to observe effects that have been previously studied only under the assumption of having a material available with a dispersive permeability. Moreover, all-dielectric nanostructures do not suffer from dissipation, i.e., nonradiative losses, as it would have happened in comparable structures made from metals.[130,131] This spurred quite a lot of attention since the total efficiency is decisive for the translation from a scientific finding to an innovation. Also hybrid antennas that combine the benefits of metallic structures, i.e., strong field localization, and dielectric structures, i.e., good control over the directional emission thanks to the spatial interference of multiple multipolar moments, are an important field of research.[132]

In a second important research strand, the multipole expansion of photonic nanostructures with nonlinear responses attracted quite a lot of attention as it offers interesting insights into the necessary design.[133–137] For example, it has been appreciated that for an optimal second harmonic conversion a double resonant antennas is useful, i.e., a nanoantenna supporting a resonance at both the fundamental and the second harmonic frequency.[138] Moreover, the induced modes should couple well to free space radiation to avoid a quenching of the generated second harmonic as well as a strong excitation with free space radiation. This prompts to design the nanoantennas to support strong electric dipole moments. Finally, also a good modal overlap is required between the induced nonlinear polarization and the modes sustained at the second harmonic frequency, which prompts for a nanoantenna that supports electric quadrupole/magnetic dipole moments.[139]

In addition, the study of nonlinear effects recently got a new twist by considering all-dielectric materials to avoid the disadvantage of metal based structures.[140] The exploitation of well pronounced resonance of modes with different multipolar order at both the fundamental and, e.g., the second harmonic frequency, was shown to be decisive to perceive structures with an excellent nonlinear conversion efficiency.[141] On the other hand, the purposeful suppression of an electric dipole moment can also be used to enhance the second harmonic signal since radiation is suppressed and resonances with very large quality factors are encountered.[142] Besides, not just the efficiency is important but also the radiative characteristics of light at a higher harmonic have been studied on the base of considering the induced multipole moments.[143–145] All these properties can be well discussed on the base of the expressions summarized in this contribution. Therefore, it is important to note that our expressions can be used to obtain the nonlinear induced multipole moments by using the induced nonlinear polarizations (or induced currents).

4. Outlook

The fact that the new expressions are exact and simple can benefit applications in several different fields involving light–matter interaction. We sketch a few examples.

As exemplified in Section 3, and already exploited in the literature,[76,121–124] one application of the expressions is in the postprocessing of the currents induced in illuminated structures obtained from numerical Maxwell solvers. The accuracy of the derived predictions, for example the cross-sections, will outperform those obtained using long wavelength approximations of the multipolar moments.

Another application can be found in computational electromagnetism.
dipole approximation (DDA),\(^{[146]}\) the electromagnetic response of a complex scatterer is computed by first discretizing its volume into small elements. The electric dipole induced in each element by the external fields and the fields scattered by all the other elements is then a part of the self-consistent system of equations that provides the final solution. Evolutions of DDA that additionally use magnetic dipoles and electric quadrupoles have been developed as well.\(^{[147]}\) The long wavelength approximations of the multipolar moments are used in these methods. Their upgrade with the exact expressions would result in either increased accuracy for given computational resources, or in computational savings for a given target accuracy due to the ability to use larger discretization volumes.

The long wavelength approximation of multipoles is often used in the Lagrangian and Hamiltonian description of the interaction of electromagnetic fields with nuclei, atoms and molecules (refs. [91], Section IV.C.3, Equations (C31)) and\(^{[148]}\), Chap. 13). The coupling between the matter systems and the field is determined by multipolar operators. The matrix elements of such operators between initial and final states of the matter system are used to study the interaction and to predict observable quantities in nuclear, atomic, and molecular spectroscopy. The substitution of the approximated expressions by the exact ones, for example in computer codes used for the calculations of molecular responses, would improve the quality of the predictions.

A possible extension of the results to inhomogeneous media, like a particle on top of a substrate, may be done as follows. First, the induced current densities inside the particle (i.e., \(J_\omega(r)\)) and \(M_\omega(r)\) should be calculated, typically by numerical approaches. Then, the induced multipole moments can be obtained by using our exact expressions. The final electromagnetic fields produced in the inhomogeneous media (multilayer) by each multipolar term can then be obtained by expressing the Green’s tensor of the multilayer in the multipolar basis. The only difference compared to the homogeneous case is that the Green’s tensor of the multilayer is not diagonal in the multipolar basis, and each of the multipolar terms in the current will give rise to a linear combination of radiated multipolar fields.\(^{[87]}\)

5. Conclusion

We have summarized recent contributions to the theory of multipolar decompositions of localized electric current density distributions. We have also extended those results to intrinsic magnetic current densities due to spin, and derived the conditions that electromagnetic sources of pure handedness need to meet. We provide scripts that facilitate the automatic calculation of multipole moments of arbitrary order. The scripts are available online at https://github.com/RasoulAlaei/exact_multipoles. We believe that the new expressions for the multipolar moments and their automatic use will be helpful in theoretical and practical tasks in the scientific areas that use electromagnetic multipolar decompositions.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

light–matter interactions, multipole moments, nanophotonics, optical antennas, theory

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Making these substitutions in Equation (33) and recombining all the $\mathbb{P}_{\mathbf{a}}^{\mathbb{M}}(r,0)$ terms together readily leads to Equation (34).

Recalling that $\langle \mathbf{A} \rangle = \mathbf{A} \times \mathbf{r}$ is equivalent to $\mathbf{p} \times \mathbf{r}$ in $\mathbf{r}$-space, it is clear by comparing Equations (7) and (25) that the electric multipoles of $\mathbb{J}_{\mathbf{a}}^{(0)}$ are the magnetic multipoles of $\langle \mathbf{r} \rangle$, $\mathbf{r}$, and vice versa.

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