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An electromagnetic multipole expansion beyond the long-wavelength approximation

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ABSTRACT

The multipole expansion is a key tool in the study of light–matter interactions. All the information about the radiation of and coupling to electromagnetic fields of a given charge-density distribution is condensed into few numbers: The multipole moments of the source. These numbers are frequently computed with expressions obtained after the long-wavelength approximation. Here, we derive exact expressions for the multipole moments of dynamic sources that resemble in their simplicity their approximate counterparts. We validate our new expressions against analytical results for a spherical source, and then use them to calculate the induced moments for some selected sources with a non-trivial shape. The comparison of the results to those obtained with approximate expressions shows a considerable disagreement even for sources of subwavelength size. Our expressions are relevant for any scientific area dealing with the interaction between the electromagnetic field and material systems.

1. Introduction

The multipolar decomposition of a given charge–current distribution is taught in every undergraduate course in physics. The resulting set of numbers are called the multipolar moments. They are classified according to their order, i.e. dipoles, quadrupoles etc. For each order, there are electric and magnetic multipolar moments. Each multipolar moment is uniquely connected to a corresponding multipolar field. Their importance stems from the fact that the multipolar moments of a charge–current distribution 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 any scientific area dealing with the interaction between the electromagnetic field and material systems. In particle physics, the multipole moments of the nuclei provide information on the distribution of charges inside the nucleus. In chemistry, the dipole and quadrupolar polarizabilities of a molecule determine most of its properties. In electrical engineering, the multipole expansion is used to quantify the radiation from antennas. And the list goes on.

In this contribution, we present new exact expressions for the multipolar decomposition of an electric charge–current distribution. They provide a straightforward path for upgrading analytical and numerical models currently using the long-wavelength approximation. After the upgrade, the models become exact. The expressions that we provide are directly applicable to the many areas where the multipole decomposition of electrical current density distributions is used. For the sake of concreteness, in this article we apply them to a specific field: Nanophotonics.

In nanophotonics, one purpose is to control and manipulate light on the nanoscale. Plasmonic or high-index dielectric nanoparticles are frequently used for this purpose [1,2]. The multipole expansion provides insight into several optical phenomena, such as Fano resonances [3,4], electromagnetically-induced-transparency [5], directional light emission [6–11], manipulating and controlling spontaneous emission [12–14], light perfect absorption [15–17], electromagnetic cloaking [18,19], and optical (pulling, pushing, and lateral) forces [20–24]. In all these cases, an external field induces displacement or conductive currents into the particles. These induced currents are the source of the scattered field. But: How can we calculate the multipole moments of these induced current distributions?

Exact expressions exists and can be found in standard textbooks, e.g. Eq. (7.20) in [25] or Eq. (9.165) in [26] (without the magnetization current therein) and a new formulation have been recently derived

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in [27]. However, up to now they are not frequently used in the literature. One reason for this may be their complexity, i.e. they feature differential operators like the curl and/or vector spherical harmonics. Instead, a long-wavelength approximation that considerably simplifies the expressions is very often used in nanophotonics [28–34]. Their integrands contain algebraic functions of the coordinate and current density vectors. Moreover, the approximate expressions resemble those for the multipole moments derived in the context of electro-statics and magneto-statics. To set a starting point, these expressions are documented in Table 1. The so-called toroidal moments are also included in these expression as the second term in the electric multipole moments [30,34,35]. It is important to mention that there is an alternative approach to calculate the multipole moments which is based on the scattered fields [26,36,37]. They are exact and valid for any particle's size. We note that the multipole moments, just as any other quantity in physics, have identical physical meaning independent on their basis (Cartesian or spherical) or which approaches (scattered fields or induced currents) has been used to extract them. The change of basis (Cartesian to spherical and vise versa) will not change the physical meaning of the multipole moments (see the *supplementary material* for the relation between two basis).

2. Derivation of the multipole moments

Let us first investigate the range of validity of the expressions in Table 1 by comparing them with Mie theory. In Mie theory, the solution for the scattering of a plane wave by a sphere is obtained without *any approximation*, i.e. it is valid for *any wavelength and size of the sphere*. For example, Mie theory allows to compute the individual contributions of each induced electric and magnetic multipole moment to the total scattering cross-section. We will compare those exact individual contributions to the ones obtained using the formulas in Table 1. We consider a high-index dielectric nanosphere and a gold nanosphere. Both are illuminated with a linearly x-polarized plane wave that propagates in the z-direction. The induced multipole moments in both cases can be computed using the expressions in Table 1. The induced electric current density is obtained by using $J_\omega(\mathbf{r}) = i\omega\epsilon_0\left(\epsilon_r - 1\right)\mathrm{E}_\omega(\mathbf{r})$, where $\mathrm{E}_\omega(\mathbf{r})$ is the electric field distribution, ϵ_0 is the permittivity of free space, and ϵ_r is the relative permittivity of the sphere. The permittivity of the dielectric sphere is assumed to be $\epsilon_r = 2.5^2$. Dispersive material properties as documented in the literature are considered for gold [38]. We assume air as the host medium. We used a numerical finite element solver to obtain the electric field distributions [39].

Using the multipole moments, it is easy to obtain the total scattering cross section, i.e. the sum of the contributions from different multipole moments, as [26]:

$$\begin{aligned} C_{\mathrm{sca}}^{\mathrm{total}} &= C_{\mathrm{sca}}^{p} + C_{\mathrm{sca}}^{m} + C_{\mathrm{sca}}^{Q^e} + C_{\mathrm{sca}}^{Q^m} + \cdots \\ &= \frac{k^4}{6\pi\epsilon_0^2|\mathbf{E}_{\mathrm{inc}}|^2}\left[\sum_\alpha\left(|p_\alpha|^2 + \frac{|m_\alpha|^2}{c}\right) + \right. \\ &\left. \frac{1}{120}\sum_{\alpha\beta}\left(\left|kQ^e_{\alpha\beta}\right|^2 + \left|\frac{kQ^m_{\alpha\beta}}{c}\right|^2\right) + \cdots\right] \end{aligned} \tag{1}$$

where, p_α, m_α are the electric and magnetic dipole moments, respectively. $Q^e_{\alpha\beta}$, $Q^m_{\alpha\beta}$ are the electric and magnetic quadrupole moments, respectively. $|\mathbf{E}_{\mathrm{inc}}|$ is the electric field amplitude of the incident plane wave, k is the wavenumber, and c is the speed of light.

Fig. 1 shows the contribution of each multipole moment to the scattering cross section for a high-index dielectric as well as a gold nanosphere. The results obtained using the approximate expression are compared with those obtained from Mie theory. It can be seen that, upon increasing the a/λ ratio, there is a large deviation between the scattering cross section obtained from the expressions in Table 1 and the Mie theory. The relative error between the two approaches is shown in Fig. 1(c) and (d). The relative error is more than 100% for the dielectric sphere at $2a/\lambda \approx 0.75$ for both electric and magnetic dipole moments. This large deviation occurs because the expressions in Table 1 are obtained in the long-wavelength approximation [26], i.e. they are only valid for particles small compared to the wavelength of the incident light (i.e. $D \ll \lambda$ where D is the biggest dimension of the particle).

Thus, the long-wavelength expressions in Table 1 can *not* be used for large particles (compared to the wavelength). The large deviation observed in Fig. 1(c) and (d) for different multipole moments will significantly affect the quantitative prediction of *multipolar interference*, which is the main physical mechanism behind Fano resonances [3,4], directional light emission [8–11], and light perfect absorption [15,16]. Moreover, any physical quantity obtained using the multipole moments of Table 1, e.g. absorption/extinction cross section, or optical torque/force, carries a corresponding error. Therefore, the application of the exact expressions for the multipole moments is important since it provides a better understanding of all the highlighted optical phenomena and enables its quantitative prediction.

To improve the situation and indeed to provide error-free expressions, we now derive exact expressions for the induced electric and magnetic multipole moments that are valid for *any wavelength and size* (see Table 2). They can be used to compute the multipole moments of arbitrarily shaped particles. Our exact expressions for multipole moments are very similar to the well-known expression obtained in long-wavelength approximation (see Table 1).

Our starting point are the hybrid integrals in Fourier and coordinate space in Eq. 14 of [35] (see the *supplementary material*). These integrals are exact expressions for all the multipolar moments of a spatially confined electric current density distribution. They are valid for any size of the distribution. Crucially, the Fourier space part of the integrals does not depend on the current density. The results in Table 2 are obtained after carrying out the Fourier space integrals for the electric and magnetic dipolar and quadrupolar orders (see the *supplementary material*). Our results have two main advantages with respect to other exact expressions [25–27]. One is that our formulas are simpler: The previously existing expressions contain differential operators and/or vector spherical harmonics inside the integrands, while ours contain algebraic functions of the coordinate and current density vectors, and spherical Bessel functions. The other advantage is that the previous expressions lack the similarity to their long-wavelength approximations that ours have (compare Tables 1 and 2). Therefor, our expressions allow a straightforward upgrade of analytical and numerical models using the approximated long-wavelength expressions. After the upgrade, the models become exact.

Basically, any code that has been previously implemented to compute the multipole moments with the approximate expression can be made to be accurate with a marginal change.

In order to show the correctness of the expressions in Table 2, we compute the contributions of different multipole moments to the scattering cross section and compare them to those obtained with Mie theory. Fig. 2 shows the different contributions as a function of the particle's size parameter $2a/\lambda$ for both the previously considered dielectric and gold spheres. It can be seen that the results from our exact expressions are in excellent agreement with those from Mie theory, irrespective of the particle's size parameter. Indeed, they are indistinguishable up to a numerical noise level.

Up to now, we have considered only spherical particles that could also be studied with Mie theory. We now use the new expressions in Table 2 to calculate the induced moments of a canonical particle made of two coupled nanopatches. Its geometry and the results are shown in Fig. 3. The coupled nanopatches support a strong electric and magnetic response. The radius and thickness of the coupled disk is assumed to be $a = 250$ nm, $t = 80$ nm, respectively. The spacer between the two disks is $g = 120$ nm. It can be seen that there is a significant deviation between the contributions to the scattering cross section from the different multipole moments as predicted by the approximate (Table 1) and by the exact (Table 2) expressions. The relative error is shown in Fig. 3(b). Some of them reach 25% for a particle size of about half the wavelength.