

An Algebraic Approach to Light–Matter Interactions

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Abstract: A theoretical and computational framework for the study and engineering of light–matter interactions is reviewed in here. The framework rests on the invariance properties of electromagnetism, and is formalized in a Hilbert space whose conformally invariant scalar product provides connections to physical quantities, such as the energy or momentum of a given field, or the outcome of measurements. The light–matter interaction is modeled by the polychromatic scattering operator, which establishes a natural connection to a popular computational formalism, the transition matrix, or T-matrix. This review contains a succinct yet comprehensive description of the main theoretical ideas, and illustrates some of the practical benefits of the approach.

1. Prologue

From nano physics to cosmology, and from computer chips to radiation therapy, Maxwell equations are central in our scientific and technological development. The more we master the theory and engineering of light–matter interactions, the deeper we understand fundamental physics, and the better the technologies that we develop. New fabrication techniques developed in the last few decades, such as for example,^[1–4] feature an exquisite control of the microscale and therewith provide us with many exciting design possibilities. At the same time, substantial theoretical and computational challenges arise regarding the optimal exploitation of such new capabilities. In this context, the development of theoretical and computational tools with increasing generality and efficiency is important.

This review explains recent developments^[5–11] that have taken a theory of free fields, extended it for modeling linear light–matter interactions, and connected the extended theory to a popular computational approach by upgrading the latter from its typical monochromatic setting to a general polychromatic one. The theory is based on invariance properties of electromagnetism, which guide the formalism in a Hilbert space. The Hilbert space is the

arena where the physical effects of light–matter interaction are modeled using the scattering operator, and where they become computationally accessible. The algebraic setting affords a compact notation which facilitates theoretical developments and computer implementation. Examples of such computations are scattering, absorption, and the changes in the fundamental quantities of the field upon interaction with matter, such as energy or momentum. The vast toolbox of matrix algebra becomes available for the study and engineering of light–matter interactions. For example, the generalized eigenvalue decomposition of two Hermitian

matrices can be used to readily obtain the pulse of light that exerts the maximum torque per unit of energy onto a given object. The setting provides a unified framework where the same fundamental tools can be effectively applied to diverse questions, which are currently addressed using diverse methodologies, such as calculation and optimization of optical forces and optical torques,^[12–15] faithful simulation of single photon fields,^[16,17] computation of light–matter interaction at relativistic speeds,^[18–20] quantification of symmetry breaking,^[21–28] including chirality,^[29–36] and the study of optical helicity and its corresponding electromagnetic duality symmetry.^[37–58] Some theoretical and computational limitations and challenges are identified as targets for further research, such as the treatment of quantum light and non-linear light–matter interactions. In the view of the present author, it is worth exploring the limits of this algebraic approach: The general theoretical basis and the compact notation allow one to conveniently develop new theoretical ideas, which can be readily implemented and tested numerically, thanks to the tight connections to popular computational tools.

2. Introduction: Context and Summary

During the first decades of the twentieth century, Einstein brought the concepts of symmetry and invariance to the forefront of theoretical physics. First with the special theory of relativity, and later with the general theory of relativity. According to Gross^[59] “Einstein’s great advance in 1905 was to put symmetry first, to regard the symmetry principle as the primary feature of nature that constrains the allowable dynamical laws.”

In 1939, Wigner established the foundations of the standard model with a simple yet powerful idea based on invariance: A particle is an object with properties such as mass and spin that take the same values in all inertial reference frames, that is, under all the transformations of the Poincaré group of special relativity. Such group is composed by space and time translations, spatial rotations, and Lorentz boosts. The latter are

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transformations between inertial reference frames. In particular, Wigner defined a photon as an object with zero mass and helicity +1 or -1, which correspond to left or right circular polarization handedness, respectively. Such values fix the way in which a photon will transform under the Poincaré group by fixing what is known as a unitary irreducible representation of such group, that is, an elementary representation [60, Chap. 10].

The seminal work about the invariance of Maxwell equations and of the speed of light under Lorentz boosts had been done by Lorentz himself in 1904,[61] and it was understood very soon thereafter that the Poincaré group does not exhaust the invariance of electromagnetism. In 1910, Bateman and Cunningham showed that the largest group of transformations that leave Maxwell equations with sources invariant is the fifteen-parameter conformal group in 3+1 Minkowski spacetime.[62,63] The Poincaré group is a subgroup of such conformal group. The same result was also obtained later by Dirac.[64] That sources are included in the invariance deserves further discussion. Even though the conformal invariance of Maxwell equations with sources is often explicitly recognized in the literature; e.g., see [65], Section 3.1], or,[66] one may however reach the incorrect conclusion that Maxwell equations are conformally invariant only in the source-free case, because fixed mass parameters break the scale invariance. In this respect, it is important to note that Maxwell equations including electric charge-current densities $[\rho(\mathbf{r}, t), \mathbf{J}(\mathbf{r}, t)]$, and magnetization and polarization densities $[\mathbf{M}(\mathbf{r}, t), \mathbf{P}(\mathbf{r}, t)]$ do not feature any fixed mass parameter. The conformal invariance follows from the way in which the sources transform. The conformal transformations of $\rho(\mathbf{r}, t)$ and $\mathbf{J}(\mathbf{r}, t)$ can be found in ref. [65, Eqs. 3.40ab]. While in the work of Dirac[64] the sources were electric charge-current densities, both Bateman[62] and Cunningham[63] included magnetization and polarization densities as well. The bottom line is that when both fields and sources are transformed, the form of the dynamic Maxwell equations with sources remains invariant. Even for equations with fixed mass parameters, such as the Dirac equation, conformal invariance can be maintained if one allows the mass to change as well,[67] which is arguably not unlike the change of the Newtonian mass in different reference frames. Besides the Poincaré group, the conformal group contains spacetime scalings and special conformal transformations.[68] Scalings can be understood as a global change of measurement units, and the special conformal transformations as spacetime-dependent changes of measurement units.[66,68] The latter were sometimes confused with changes to accelerating reference frames. The conformal symmetry, its role in theoretical physics, and their historical evolution are comprehensively explained in the review by Kastrup.[66]

Powerful as it is, the concept of invariance is by itself far from practical use. A large step towards practicality is taken when a Hilbert space is identified. The Hilbert space of physical states, in this case electromagnetic fields, where invariance can be made explicit as the invariance of physical quantities upon transformations by the operators that represent the relevant symmetries in the Hilbert space. In his important 1964 paper,[69] Gross identified such Hilbert space for free fields, that is, for fields that are not interacting with matter. Gross showed that a scalar product that was known to be invariant under the Poincaré group is also invariant under the conformal group, and that the photonic representation of the Poincaré group identified by Wigner

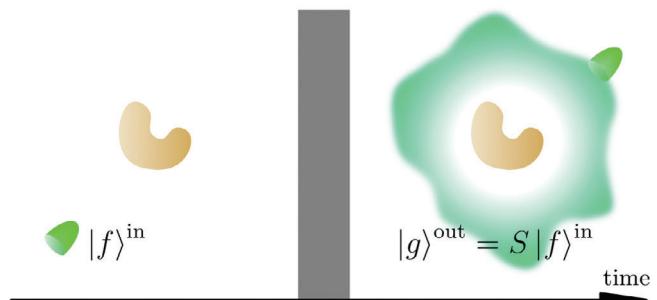


Figure 1. An electromagnetic field interacts with a material object during the grayed-out period. In the algebraic approach to light-matter interactions, the incoming field $|f\rangle^{\text{in}}$ and the outgoing field $|g\rangle^{\text{out}}$ belong to the Hilbert space of solutions of Maxwell equations \mathbb{M} . The incoming and outgoing fields are connected by the scattering operator S , which defines the action of the object on the fields.

bijectively determines an irreducible representation of the conformal group. The Hilbert space from Gross is composed by the electromagnetic fields whose norm under the scalar product is finite. These results were given an even deeper physical meaning by Zeldovich in 1965,[70] who proved that the norm squared of a given field is actually the number of photons in such field. Moreover, fundamental quantities of the field, such as energy, linear momentum, or angular momentum, can be computed by means of scalar products.[71] Several works on electromagnetic Hilbert spaces that considered the Poincaré and/or the conformal groups appeared in the late 1960s and early 1970s.[72–79] Unfortunately, continuations have been scarce.[66,71,80,81]

Even with the appropriate Hilbert space in place, the practical use of these algebraic tools for modeling light-matter interaction is not clear. One difficulty is that the scalar product and the Hilbert space apply to free fields. This difficulty is solved by the scattering operator.[82,83] The scattering operator of a given object maps the free fields before the light-matter interaction to the free fields after the light-matter interaction. There is a period which is left out: From the time when the illumination first “touches” the material object, until the time when the object has finished re-radiating. This is illustrated in **Figure 1**. The scattering operator is hence an operator in the Hilbert space of free Maxwell fields. During the excluded time period, one cannot use the scalar product to compute the number of photons, energy, or other fundamental quantities. However, it is always theoretically possible to use the scattered free fields to obtain the near fields up to the surface of the object, because the relationship between near and far fields is bijective [84, Chap. 9] [85, §§2-3, Chap. 5, Eqs. 2.12(a,b)].

Many of the elements necessary for computer implementation are now ready. The crucial connection between abstract algebra and computer calculations is made through the concept of orthogonality inherent to a Hilbert space. With orthogonality comes the ability to expand any element of the Hilbert space as a unique linear superposition of the elements of a basis set. The computer calculations are then performed using the expansion coefficients, which are typically arranged as a column vector for convenience. Operators are then represented by basis-dependent matrices, and the light-matter interaction is modeled by the multiplication of the vector representing the illumination with the matrix representing the scattering operator. The resulting column vector con-

tains the expansion coefficients of the resulting field. There is an approach to computational electromagnetism in physics and engineering that has used the matrix-vector paradigm for decades, namely the transition matrix, or T-matrix approach. The T-matrix is a popular and powerful formalism for computing light–matter interactions whose origin can be pinpointed to the seminal 1965 paper by Waterman.^[86] The T-matrix encodes the full linear electromagnetic response of a given object upon arbitrary illuminations, and is bijectively connected to the scattering operator. This provides the natural framework for the algebraic approach. In contrast to research on abstract algebra for electromagnetism, the T-matrix has enjoyed plenty of attention, as the hundreds of references collected in databases show.^[87,88]

One obstacle remained until recently, however: The T-matrix formalism was originally defined and has been developed systematically assuming that the illuminating and scattered fields are monochromatic. Monochromatic fields feature the harmonic time dependence $\exp(-i\omega_0 t)$ for some fixed frequency ω_0 , and have no beginning or end in time. It is clear that the linearity of Maxwell equations permits the computation of the response of an object to any given polychromatic illumination by superposing the responses to many monochromatic fields with different frequencies,^[89] and there exist general works on time-domain scattering.^[90,91] However, a systematic development of the polychromatic T-matrix did not exist, which in particular prevented the direct treatment of the interaction of objects with light pulses, and the computation of light–matter interactions for objects moving at constant speeds with respect to the source and/or measurement device. While the source could be approximately monochromatic in its frame, new frequencies appear after considering the light in the reference frame of the moving object. The remaining obstacle has been recently removed by Maxim Vavilin and the present author,^[10] by following the guidelines provided by Wigner in the Poincaré group to generalize the usual monochromatic formalism to the case of polychromatic light–matter interaction. Following Wigner forces one to consider polychromatic fields from the start because Lorentz boosts change the frequency and therefore render any monochromatic approach immediately obsolete. Group theory had already been used for extending the applicability of the T-matrix. Waterman's single-object formalism^[86] was extended to multiple coupled objects by Peterson and Ström.^[92] They used the theory of the 3D Euclidean group, which is sufficient for the monochromatic case, for conveniently formulating the translations and rotations of individual T-matrices that are needed when computing the T-matrix of the composite object.

The computational convenience of the framework, mostly due to its tight connection to the T-matrix formalism, deserves to be highlighted. The electromagnetic response of macroscopic objects, such as silicon disks or metallic helices can be modeled by their T-matrices. The same is true for microscopic objects such as molecules.^[93] The T-matrix affords numerically efficient computations of the response of composite objects,^[94–96] in particular for periodic arrangements^[97–99] such as metasurfaces. It seems clear that the T-matrix will keep attracting attention because of its generality and computational efficiency. The formalism has been developed and improved over many decades, resulting in a very substantial body of literature,^[87,88] and a continuously growing list of publicly available computer codes^[100,101] produced by a vibrant

research community. One salient advantage of publicly available open-source codes is that they make reproducibility and scrutiny much easier than when closed source software is used instead. The recently released `treams` Python package,^[99,102,103] which is publicly available at <https://github.com/tpf-photronics/treams>, has been used in some of the works reviewed here. Such articles provide all the extra python code necessary to reproduce their results. In effect, `treams` provides the basis for implementing the computational side of the algebraic approach.

In addition to computational convenience, the algebraic framework also provides tools and guidance for deriving new results, which sometimes lead to new research questions, and to connections to other fields of physics. For example, an extension of the formalism to matter has lead to the identification of a new scalar product for computing fundamental quantities in static matter,^[9] and to showing that the electromagnetic helicity of the free electromagnetic field and the screwiness of the static magnetization density in matter are two embodiments of the same quantity.^[8] The connection establishes the theoretical basis for studying the conversion between the two embodiments, and motivates its further theoretical, numerical and experimental investigation. In particular, because the said screwiness of the static magnetization density is essentially the magnetic helicity,^[104–106] a concept relevant in diverse areas of physics such as cosmology,^[107,108] solar physics,^[109] fusion physics,^[110,111] magneto-hydrodynamics,^[112–115] and condensed matter.^[116–120]

The rest of the article is organized as follows. The electromagnetic Hilbert space and scalar product are reviewed in Section 3. The members of the Hilbert space are polychromatic fields with a finite number of photons and finite energy. The latter condition ensures that incoming fields finish and outgoing fields start, which make them perfectly suited for modeling absorption and emission from matter, respectively. Some emphasis is placed on the conventions that are used for integrals in the frequency and momentum spaces, and in the definitions of basis vectors such as plane waves and multipolar fields.^[10] Such conventions have several benefits. One is that both the basis vectors and the expansion coefficient functions transform unitarily under the Poincaré group, following the prescription from Wigner. This means in particular that they transform unitarily under changes of inertial reference frame, which significantly facilitates the computation of the interaction of electromagnetic pulses with material objects moving at relativistic speeds with respect to the source of light.^[121] Another benefit is the extension of the scalar product to outgoing and incoming fields, that is, fields with outwards and inwards fluxes at spatial infinity, respectively. Such extension is necessary because typical expressions for the scalar product apply to stationary fields with zero net flux at spatial infinity. A third benefit is the simplicity of the expressions for the scalar product as a function of the expansion coefficient functions corresponding to plane waves and multipoles. On the one hand, such simplicity facilitates the implementation of the expressions in computer codes, and on the other hand, the conventions result in units of meters for such coefficient functions, which eases the consistency checks during analytical derivations. Emphasis is also placed on two important uses of the scalar product. One is the computation of fundamental quantities of a given field, such as energy, helicity or momentum, by “sandwiching” the corresponding self-adjoint operator representing the quantity between the

bra and ket versions of the field. The other is the consistent modeling of physical measurements,^[9] whose results are required to be invariant under all the allowed changes of reference frame, which in this case means under all the transformations in the conformal group.

Section 4 reviews two connections between the algebraic formalism and popular computational strategies in light-matter interaction: The T-matrix, and generic Maxwell solvers such as COMSOL, MEEP, JCMsuite, CST, or Lumerical. Section 4.1 explains how the T-matrix is the natural computational complement of the abstract algebraic formalism. The power of this connection is shown by reviewing an example where the optical force exerted by a light pulse onto a silicon sphere is computed for a wide range of relativistic speeds of the sphere.^[121] The possibility to write down and solve optimization problems is discussed, as for example, for finding out the most energy efficient polychromatic field for exerting torque on a given object. Section 4.2 reviews a recent result that allows one to compute scalar products between pairs of outgoing or incoming fields using only the values of the fields on a closed spatial surface.^[11] The capabilities of popular Maxwell solvers are thereby augmented, for example, by a robust and simple method for normalizing emitted fields so that they contain a single photon.

The convenient and effective way in which symmetries and selection rules can be treated in the Hilbert space is reviewed in Section 5. In particular, selection rules arise because the interaction of light with a symmetric object does not couple the electromagnetic eigenstates of the symmetry with different eigenvalue. The selection rules that control the coupling of electric or magnetic emitters onto waveguides are presented as an example. Besides selection rules that apply when the object is symmetric, the definitions of quantitative measures of symmetry breaking by a given object,^[6] and the electromagnetic chirality of a given object^[45,122] are also discussed.

Section 6 reviews the extension of the algebraic approach to matter in the sense that fundamental quantities of static matter can be computed using scalar products.^[9] Static matter is represented by its electric charge $\rho(\mathbf{r})$ and magnetization $\mathbf{M}(\mathbf{r})$ densities, and an appropriate scalar product and group of invariance are reviewed. The group of invariance turns out to be also a conformal group, but this time for the Euclidean space with three spatial dimensions, instead of the 3+1 Minkowski spacetime. The possibility of extending definitions of fundamental quantities that exist for the fields onto matter is exemplified for the case of the helicity operator in Section 6.1. This particular extension allows one to define helicity in matter, a possibility that has been debated in the literature. The definition that one obtains in this way, which can be seen as a measure of the degree of twistedness of the static magnetization, is essentially equivalent to the magnetic helicity, a well-known quantity relevant in many fields of physics.

Finally, Section 7 concludes the article with a discussion about some immediate applications of the formalism, and some future research directions.

3. The Hilbert Space

Figure 1 depicts a light-matter interaction sequence. An incoming electromagnetic field approaches a material object. Then, the

field and the object interact for a finite period of time, gray in the figure. After the interaction has finished, there is a resulting outgoing field. Incoming and outgoing fields feature inwards and outwards fluxes at spatial infinity, respectively.

Before the field reaches the object and after all the re-radiation by the object has finished, the electromagnetic fields are free, that is, they are solutions of Maxwell equations without sources. The set of such solutions is the vector space that, together with the appropriate scalar product, constitute our Hilbert space \mathbb{M} . Section 3.1 is dedicated to the scalar product, and highlights its useful connections to physical quantities. For example, with such scalar product, the norm squared of $|f\rangle$, $\langle f|f\rangle$, is the number of photons in the field. This was shown by Zeldovich in 1965.^[70] Since all the members of a Hilbert space must have finite norms, it follows that the members of \mathbb{M} have finite numbers of photons. A finite energy must also be required on physical grounds.

In this setting, the effect of a given object on the fields is modeled by a linear operator S , which maps any incoming field $|f\rangle^{\text{in}}$ into its corresponding outgoing field $|g\rangle^{\text{out}}$:

$$|g\rangle^{\text{out}} = S |f\rangle^{\text{in}} \quad (1)$$

Symmetries and their generators are also represented by operators that act on the states in \mathbb{M} , and map them back to \mathbb{M} . The generators of continuous symmetries represent fundamental quantities of the electromagnetic field, such as energy or angular momentum. Section 5 reviews how symmetries and their consequences are formalized in \mathbb{M} .

The multipolar fields, also known as spherical waves, provide a convenient way of expanding the incoming and outgoing fields of Figure 1:

$$\mathbf{E}(\mathbf{r}, t)^{\text{in/out}} = \sum_{\lambda=\pm 1} \sum_{j=1}^{\infty} \sum_{m=-j}^j \int_{>0}^{\infty} dk k f_{jm\lambda}(k) |kjm\lambda\rangle^{\text{in/out}} \quad (2)$$

where the $f_{jm\lambda}(k)$ are scalar complex functions, and the incoming and outgoing multipoles $|kjm\lambda\rangle^{\text{in/out}}$ are defined as [10, Eq. (92)]:

$$|kjm\lambda\rangle^{\text{in/out}} \equiv -\frac{1}{2} \sqrt{\frac{c_0 \hbar}{\epsilon_0}} \frac{1}{\sqrt{2\pi}} k i^j \exp(-ikc_0 t) \times \left[\mathbf{N}_{jm}^{\text{in/out}}(k|\mathbf{r}|, \hat{\mathbf{r}}) + \lambda \mathbf{M}_{jm}^{\text{in/out}}(k|\mathbf{r}|, \mathbf{r}) \right] \quad (3)$$

where c_0 is the speed of light, \hbar the reduced Planck constant, ϵ_0 the permittivity of vacuum, $k = |\mathbf{k}| = \omega/c_0$ is the wavenumber, and the \mathbf{N} and \mathbf{M} are the usual “electric” and “magnetic” multipoles (see e.g., [123, Sec. 9.7], [10, Eqs. (50,51)]). SI units will be used throughout the article. The incoming or outgoing character of the multipoles is determined by the kind of the spherical Hankel functions contained in \mathbf{N} and \mathbf{M} : The first kind $h_l^1(kr)$ defines outgoing waves, and the second kind $h_l^2(kr)$ incoming waves. The reason for using the integration measure $dk k$ in Equation (2) is explained at the end of this section.

The electric field in Equation (2) is complex-valued since only positive frequencies $\omega = kc_0 > 0$ are included. The negative fre-

quencies do not contain additional information. The corresponding real-valued field can be obtained as:

$$\mathcal{E}(\mathbf{r}, t) = 2\mathbb{R}\{\mathbf{E}(\mathbf{r}, t)\} \quad (4)$$

This relationship is readily obtained by taking $\mathbf{E}(\mathbf{r}, t)$ to be the inverse Fourier transform of the positive-frequency spectrum of $\mathcal{E}(\mathbf{r}, t)$.

The $|kjm\lambda\rangle^{\text{in/out}}$ are eigenstates of four operators, the energy H , the total angular momentum squared $J^2 = J_x^2 + J_y^2 + J_z^2$, the angular momentum along the z -axis J_z , and the helicity Λ :

$$\begin{aligned} H|kjm\lambda\rangle^{\text{in/out}} &= \hbar k c_0 |kjm\lambda\rangle^{\text{in/out}}, \\ J^2|kjm\lambda\rangle^{\text{in/out}} &= \hbar^2 j(j+1) |kjm\lambda\rangle^{\text{in/out}}, \\ J_z|kjm\lambda\rangle^{\text{in/out}} &= \hbar m |kjm\lambda\rangle^{\text{in/out}}, \text{ and} \\ \Lambda|kjm\lambda\rangle^{\text{in/out}} &= \hbar\lambda |kjm\lambda\rangle^{\text{in/out}} \end{aligned} \quad (5)$$

where $j = 1$ corresponds to the dipoles, $j = 2$ to the quadrupoles, and so on, $m = -j, -j+1, \dots, j-1, j$, and $\lambda = +1(-1)$ corresponds to left(right) polarization handedness: The helicity of the field.^[71,124–130] The helicity operator is the projection of the angular momentum vector onto the direction of the linear momentum vector [60, Eq. 8.4-5]:

$$\Lambda = \frac{\mathbf{J} \cdot \mathbf{P}}{|\mathbf{P}|}. \quad (6)$$

Recent works reveal the benefits of using helicity for describing polarization, in particular at microscopic scales.^[37–46,49,50,52–56]

The choice of helicity multipoles instead of the more common parity multipoles carries theoretical and practical advantages in some cases. A salient advantage is that the helicity multipoles contain a single polarization handedness, also in the near field. Actually, the helicity multipoles defined in Equation (3), split the electromagnetic field into its left and right circular polarization handedness for $\lambda = +1$ and $\lambda = -1$, respectively. This is a property of all the eigenstates of the helicity operator, which can generally be represented by the $\mathbf{F}_\lambda(\mathbf{r}, t)$ fields defined in Equation (21). Such separation holds generally for any kind of field, far fields, modal fields, and near fields, which is particularly useful for describing the handedness of light nearby microstructures. Another difference with respect to parity eigenstates, is that helicity eigenstates do not mix upon the transformations of the Poincaré group of special relativity. That is, a left-handed field remains a left-handed field in any inertial reference frame. Actually, helicity eigenstates do not mix upon the transformations of a bigger group, the conformal group, which will be reviewed later. The separation of helicity eigenstates simplifies many expressions and avoids having to consider mixing rules, as for example, when a mere translation of the spatial origin of coordinates produces fields whose parity differs from the original one. It is worth mentioning that this separation does produce orthogonal functions with respect to the dot product in \mathbb{C}^3 , that is, for example $[\mathbf{N}_{jm}(k|\mathbf{r}|, \hat{\mathbf{r}}) + \mathbf{M}_{jm}(k|\mathbf{r}|, \hat{\mathbf{r}})]^\dagger [\mathbf{N}_{jm}(k|\mathbf{r}|, \hat{\mathbf{r}}) - \mathbf{M}_{jm}(k|\mathbf{r}|, \hat{\mathbf{r}})]$ is not zero in general.

When the parity multipoles are needed, they can be obtained as the linear combinations

$$|kjm\tau\rangle = \frac{1}{\sqrt{2}} (|kjm\lambda = +1\rangle + \tau |kjm\lambda = -1\rangle), \quad (7)$$

with $\tau = 1$ for the “electric” kind and $\tau = -1$ for the “magnetic” kind.

For Equation (2) to have the common meaning of an expansion in a Hilbert space, we should have that $\langle \lambda m j k | f \rangle = f_{jm\lambda}(k)$, which then results in the condition:

$$\langle \lambda m j k | q \bar{m} \bar{\lambda} \rangle = \delta_{jj} \delta_{mm} \delta_{\lambda\bar{\lambda}} \frac{\delta(k - q)}{k} \quad (8)$$

where δ_{ab} are Kronecker deltas and $\delta(k - q)$ a Dirac delta. The latter implies that the norm of any given $|kjm\lambda\rangle^{\text{in/out}}$ diverges and is not well-defined. And this means that they do not fit in \mathbb{M} , since all the members of a Hilbert space must have finite norms. The origin of the divergence is that the $|kjm\lambda\rangle^{\text{in/out}}$ are monochromatic fields, and divergences appear as soon as monochromaticity is assumed.^[131] Incidentally, this means that they have no beginning or end: They are eternal. In reality, monochromatic fields do not exist, and, in particular, they are not suitable for describing the sequence in Figure 1 because there are no outgoing fields before the interaction, and there are no incoming fields after the interaction. While all the members of \mathbb{M} are polychromatic, their spectra can, however, be very localized around a given frequency. The fact that the $|kjm\lambda\rangle^{\text{in/out}}$ do not belong in \mathbb{M} raises two questions: Why can they be used at all, and what are their benefits.

The $|kjm\lambda\rangle^{\text{in/out}}$ can be used inside integrals such as the one in Equation (2) to define polychromatic fields of finite norm, where the $f_{jm\lambda}(k)$ meet that $\langle f | f \rangle < \infty$ with the expression of the scalar product in Equation (15). Additionally, the $|kjm\lambda\rangle^{\text{in/out}}$ have simple transformation properties under rotations, which makes them very useful for the study of the interaction of light with rotationally-symmetric objects. Moreover, the interaction of light with optically small objects such as molecules and nanostructures, can typically be well approximated by using only the first few multipolar orders.

Similar considerations apply to monochromatic plane and cylindrical waves, which also feature divergent norms. Both allow, however, the definition of fields in \mathbb{M} through the use of appropriate integrals. Plane waves have simple transformations properties under translations, and are beneficial for the study of translationally symmetric systems, such as metasurfaces. Cylindrical waves, also known as Bessel beams, are adapted to systems with cylindrical symmetry, in particular to situations including collimated and focused beams with high angular momentum. A very practical reason for using monochromatic fields is that non-monochromatic basis in \mathbb{M} have not yet been developed for practical use.

The integral in Equation (2) can define light pulses that either start or end in time. This can be first intuitively grasped from the fact that the exponentials $\exp(-i\omega t)$ are a complete basis for square-integrable functions of time, including those with a start or an end. More precisely, this connects with causality analysis^[132] that rely on the Paley–Wiener theorem (see e.g., Ref. [90, p. 641]) to show that, for finite energy pulses, incoming

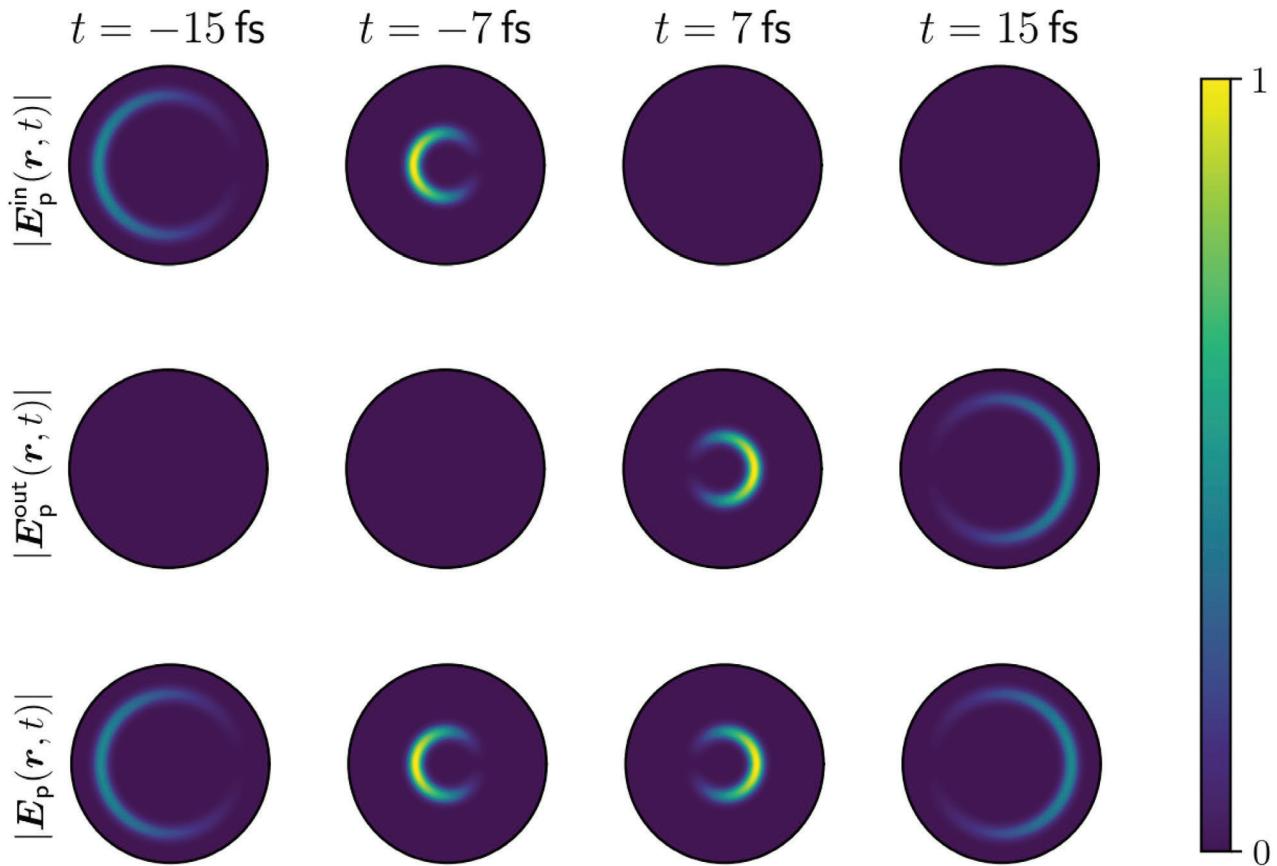


Figure 2. Adapted from Ref. [10]. Absolute values of the electric field in the zx-plane at four different points in time for the incoming, outgoing, and regular versions of the pulse in Equation (9). The z-axis points horizontally to the right, and the x-axis - vertically to the top. The incoming field in the first row is seen to be zero after some point in time, and can be thought of as having been absorbed in the singularity of the multipolar fields at the origin of coordinates. Conversely, the outgoing field in the second row is zero before some point in time, and can be thought of as being emitted from the singularity. The sum of the two, $\mathbf{E}_p(\mathbf{r}, t) = \mathbf{E}_p^{\text{in}}(\mathbf{r}, t) + \mathbf{E}_p^{\text{out}}(\mathbf{r}, t)$, produces a regular field without singularities, whose absolute value is shown in the third row.

fields have an end and outgoing fields have a beginning. A numerical illustration of this notable property can be found in Ref. [10]. **Figure 2** is a modified version of Ref. [10, Figure 1]. The first and second rows of Figure 2 depict the absolute value of the incoming and outgoing versions, respectively, of the following light pulse:

$$\mathbf{E}_p^{\text{in/out}}(\mathbf{r}, t) = A \int_{>0}^{\infty} dk k \exp\left(-\frac{(k - k_0)^2}{2\Delta^2}\right) |kj = 1 m = 1 \lambda = 1\rangle^{\text{in/out}} \quad (9)$$

where A is some normalization constant with units of length, $2\pi/k_0 = 400 \text{ nm}$, and $\Delta^{-1} = 300 \text{ nm}$.

Figure 2 shows that the incoming field in the first row is zero after some time instance, and that the outgoing field in the second row is zero before some time instance. Incoming polychromatic fields are therefore perfectly suited for modeling absorption by matter, and outgoing polychromatic fields are perfectly suited for modeling emission from matter. For example, the emission upon a particular transition between energy levels of a molecule or a quantum dot can be suitably modeled by an outgoing pulse whose total energy, as computed with Equation (16),

is equal to the difference in energy between the two levels. The shape of the pulse, in either frequency or time domain, can be adjusted to reflect theoretical expectations and/or experimental measurements of, for example, the emission linewidth.

The third row of Figure 2 corresponds to the sum of the incoming and outgoing fields $\mathbf{E}_p(\mathbf{r}, t) = \mathbf{E}_p^{\text{in}}(\mathbf{r}, t) + \mathbf{E}_p^{\text{out}}(\mathbf{r}, t)$, which is a regular field because the singularities of the spherical Hankel functions in $\mathbf{E}_p^{\text{in}}(\mathbf{r}, t)$ and $\mathbf{E}_p^{\text{out}}(\mathbf{r}, t)$ cancel each other out. Clearly, the incoming and regular fields are equal before the emission of the outgoing field starts, and the outgoing and regular fields are equal after the absorption of the incoming field has finished. The regular multipolar fields $|kjm\lambda\rangle$ are defined as in Equation (3), except that the spherical Hankel functions in \mathbf{N} and \mathbf{M} are substituted by spherical Bessel functions, and the first factor of $1/2$ on the right hand side is removed. With such definition, it holds that:

$$|kjm\lambda\rangle = |kjm\lambda\rangle^{\text{in}} + |kjm\lambda\rangle^{\text{out}} \quad (10)$$

We can use the same coefficients $f_{jm\lambda}(k)$ in a expansion such as Equation (2) and change the kind of multipolar fields to obtain the

three different behaviors, and the general relation $|f\rangle = |f\rangle^{\text{in}} + |f\rangle^{\text{out}}$.

The connection between regular, incoming, and outgoing fields overcomes an important obstacle that prevented the Hilbert space formulation of the light-matter interaction processes illustrated in Figure 1. The obstacle was that the scalar product in \mathbb{M} had been defined for regular fields without singularities.^[69] The scalar product in Ref. [69] can be written as:

$$\langle f|g\rangle = \sum_{\lambda=\pm 1} \int_{\mathbb{R}^3 - \{0\}} \frac{d^3 k}{k} f_\lambda(\mathbf{k})^* g_\lambda(\mathbf{k}) \quad (11)$$

where the complex coefficient functions are the weights of the expansion of the field into plane waves of well-defined helicity $|\mathbf{k}\lambda\rangle$:

$$E(\mathbf{r}, t) = \sum_{\lambda=\pm 1} \int_{\mathbb{R}^3 - \{0\}} \frac{d^3 k}{k} f_\lambda(\mathbf{k}) |\mathbf{k}\lambda\rangle \quad (12)$$

and the plane waves are defined as:

$$|\mathbf{k}\lambda\rangle \equiv \sqrt{\frac{c_0 \hbar}{\varepsilon_0}} \frac{1}{\sqrt{2}} \frac{1}{\sqrt{(2\pi)^3}} k \mathbf{e}_\lambda(\mathbf{k}) \exp(-ikc_0 t) \exp(i\mathbf{k} \cdot \mathbf{r}) \quad (13)$$

with $\mathbf{e}_\lambda(\mathbf{k})$ defined as in Ref. [133, Sec. 1.1.4]. The plane waves meet:

$$\begin{aligned} P_\alpha |\mathbf{k}\lambda\rangle &= \hbar k_\alpha |\mathbf{k}\lambda\rangle, \text{ for } \alpha \in \{x, y, z\}, \\ H |\mathbf{k}\lambda\rangle &= \hbar k c_0 |\mathbf{k}\lambda\rangle, \\ \Lambda |\mathbf{k}\lambda\rangle &= \hbar \lambda |\mathbf{k}\lambda\rangle, \end{aligned} \quad (14)$$

where the $P_{x,y,z}$ are the linear momentum operators.

The units of $f_{jm\lambda}(k)$ and $f_\lambda(\mathbf{k})$ are meters. Such simple units are very helpful when performing sanity checks in derivations.

The expression of the scalar product in the multipolar basis can then be readily obtained using the relationship between plane waves and regular multipoles (see e.g., Ref. [10, Eqs. (40,41)]):

$$\langle f|g\rangle = \sum_{\lambda=\pm 1} \sum_{j=1}^{\infty} \sum_{m=-j}^j \int_{>0}^{\infty} dk k f_{jm\lambda}^*(k) g_{jm\lambda}(k) \quad (15)$$

The number of photons and energy of a given field $|f\rangle$ can then be computed as:

$$\begin{aligned} \text{Number of photons } \langle f|f\rangle &= \sum_{\lambda=\pm 1} \int_{\mathbb{R}^3 - \{0\}} \frac{d^3 k}{k} |f_\lambda(\mathbf{k})|^2 \\ &= \sum_{\lambda=\pm 1} \sum_{j=1}^{\infty} \sum_{m=-j}^j \int_{>0}^{\infty} dk k |f_{jm\lambda}(k)|^2, \text{ and} \\ \text{energy } \langle f|H|f\rangle &= \sum_{\lambda=\pm 1} \int_{\mathbb{R}^3 - \{0\}} \frac{d^3 k}{k} (c_0 \hbar k) |f_\lambda(\mathbf{k})|^2 \\ &= \sum_{\lambda=\pm 1} \sum_{j=1}^{\infty} \sum_{m=-j}^j \int_{>0}^{\infty} dk k (c_0 \hbar k) |f_{jm\lambda}(k)|^2 \end{aligned} \quad (16)$$

Since, as explained above, the coefficients are the same for the regular, outgoing, and incoming versions of a field, we can extend the use of Equation (15) to incoming and outgoing fields:

$$\langle f|g\rangle =^{\text{out}} \langle f|g\rangle^{\text{out}} =^{\text{in}} \langle f|g\rangle^{\text{in}} \quad (17)$$

The extension is physically consistent, since it ensures that the value of fundamental quantities defined with Equation (19) by scalar products, such as energy $\langle f|H|f\rangle$, helicity $\langle f|\Lambda|f\rangle$, linear momentum $\langle f|P_\alpha|f\rangle$, and angular momentum $\langle f|J_\alpha|f\rangle$, is the same for the three kinds of fields when the expansion coefficients are the same. This is the case for Figure 2, where there is no light-matter interaction, in contrast to Figure 1. In the absence of interaction, the energy of the incoming field in the distant past must be equal to the energy of the outgoing field in the distant future, and also equal to the of energy of the regular field.

Additionally, Equation (17) allows one to compute scalar products between pairs of outgoing or incoming fields using their regular versions and corresponding expressions, such as Equation (15).

Before the extended discussion about the scalar product, it is worth briefly mentioning that the algebraic formalism is naturally adapted for studying and engineering the interaction of pulses of electromagnetic radiation with material objects, including scenarios where the object, the source, and the detector are moving at constant relativistic speeds with respect to each other. The origin of such capability is that the definitions in the polychromatic setting are adapted to Wigner's idea that photons transform as unitary irreducible representations of the group of special relativity, the Poincaré group. This is achieved by using the Poincaré invariant integration measure $d^3 k/k$ in expressions related to plane waves, and its result dk/k in expressions related to multipoles, together with the extra factors of k in the corresponding definitions in Equation (13) and Equation (3). Then, both the basis vectors and the corresponding coefficient functions, $f_\lambda(\mathbf{k})$ and $f_{jm\lambda}(k)$, transform unitarily under the Poincaré group in well-defined ways.^[10] The Lorentz boosts, which transform systems to different inertial reference frames, are part of the Poincaré group.

3.1. The Electromagnetic Scalar Product

A central element of any Hilbert space is its scalar product. A scalar product establishes an immediate connection to geometry, and enables crucial concepts, such as for example orthogonality: Two states are orthogonal when their scalar product vanishes. The notion of orthogonality is needed for the definition of complete orthonormal sets of vectors, that is, basis sets. The possibility of expressing any element of the Hilbert space as a unique linear superposition of the elements of a basis set, e.g., Equations (2, 12), is what connects the abstract algebra to computer calculations. Such connection is one of the strengths of the algebraic approach to light-matter interaction, as reviewed in Section 4.1.

In physics, Hilbert spaces are most commonly used in quantum mechanics. One of the key roles that scalar products play in quantum mechanics is in the definition of average values of a given observable on a given state. Another key role is in the definition of projective measurements.^[134] Both connections between physics and mathematics work also in \mathbb{M} , as follows.

Fundamental quantities of the field are represented by self-adjoint operators, such as for example energy H , or the linear momentum along the z -axis P_z . The “sandwiches” $\langle f | \Gamma | f \rangle$ are the total value of the quantity represented by Γ contained in the field [79, Chap. 3, §9]. That is, for example, the values of $\langle f | H | f \rangle$ and $\langle f | P_z | f \rangle$ coincide with the values that result from the typical integrals

$$\langle f | H | f \rangle = \frac{1}{2} \int_{\mathbb{R}^3} d\mathbf{r} \left(\epsilon_0 \mathcal{E} \cdot \mathcal{E} + \frac{1}{\mu_0} \mathcal{B} \cdot \mathcal{B} \right), \text{ and}$$

$$\langle f | P_z | f \rangle = \epsilon_0 \int_{\mathbb{R}^3} d\mathbf{r} \mathcal{E} \times \mathcal{B} \Big|_z, \text{ respectively.} \quad (18)$$

That $\langle f | \Gamma | f \rangle$ is the value of Γ in $|f\rangle$ results from the interpretation that an *observable* property is represented by a self-adjoint operator Γ , and that the value of such property in a ket is computed by the trace rule, which for a pure state $|f\rangle$ leads to the “sandwiches” in $\langle f | \Gamma | f \rangle$:

$$\text{Trace}\{\Gamma |f\rangle \langle f|\} = \langle f | \Gamma | f \rangle. \quad (19)$$

When $\Gamma = I$, the norm squared of $|f\rangle$, $\langle f | f \rangle$, is the number of photons in the field,[70] that is, the number of photons in a given normalized polychromatic mode $|\hat{f}\rangle = |f\rangle / \sqrt{\langle f | f \rangle}$. More precisely, $\langle f | f \rangle$ is the average number of photons. While, for a Fock state, $\langle f | f \rangle$ would produce an integer equal to the actual number of photons, for a coherent state it would be in general a non-integer value equal to the actual average number of photons of the coherent state. Statistical mixtures of different modes are not covered by Equation (19).

Equation 19 is valid for any self-adjoint operator mapping elements in \mathbb{M} back onto \mathbb{M} , that is, for any self-adjoint operator that maps any solution of Maxwell equations into another solution. Examples of operators that do not meet such requirement and can hence not be considered proper operators in \mathbb{M} are the position operator \mathbf{r} [71, § 14], or the spin-1 matrices.[135] In particular, the latter break the transversality of the fields [42, Chap. 3].

With respect to measurements: The invariance properties of the scalar product underpin the consistent definition of projective measurements. The outcomes of rather general electromagnetic measurement setups can be modeled as $|\langle d | f \rangle|^2$, where $|f\rangle$ is the field and $|d\rangle$ the mode of the detector. In Figure 3, $|d\rangle$ is well approximated by a plane wave. One can readily envision models for single mode fibers followed by power detectors, (chiral) spectrometers, and so on. The $|\langle d | f \rangle|^2$ are unitless numbers that represent the measurement outcome, for example the number of “clicks” in a photodetector.

A fundamental property of the scalar product in \mathbb{M} is that it is conformally invariant:[69] That is, the value of $\langle f | g \rangle$ is identical to the scalar product between $X |f\rangle$ and $X |g\rangle$, for any transformation X in the conformal group in 3+1 Minkowski spacetime, $C_{15}(3, 1)$:

$$(X |f\rangle)^\dagger (X |g\rangle) = \langle f | X^\dagger X | g \rangle = \langle f | g \rangle \quad (20)$$

The second equality is the manifestation of the invariance.

The 15-parameter conformal group consists of [65, 136]: Space-time scalings, four special conformal transformations, and the

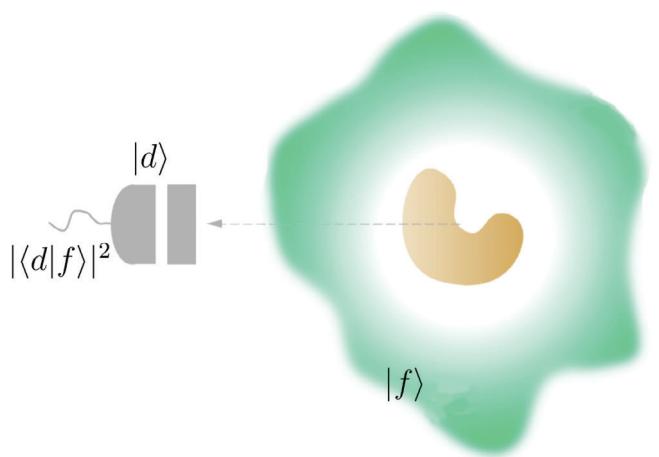


Figure 3. The outcome of an apparatus for measuring the field $|f\rangle$ can be modeled as $|\langle d | f \rangle|^2$, that is, the modulus square of the projection of the field onto the electromagnetic mode of the detector $|d\rangle$.

Poincaré group, which consists of four spacetime translations, three Lorentz boosts, and three spatial rotations. The conformal group is the largest group of invariance of Maxwell equations *including sources as spacetime densities*. Matter models such as charge densities and magnetization densities appear already in the seminal works of Bateman and Cunningham^[62,63] when they showed the conformal invariance of electrodynamics. One may then consider conformal invariance similarly as Einstein considered the special-relativistic invariance under the Poincaré group. If one imposes that measurement outcomes must be independent of the reference frame, it follows that $|\langle d | f \rangle|^2$ must be invariant under all the transformations in $C_{15}(3, 1)$, since those are valid changes of reference frame. The value of $|\langle d | f \rangle|^2$ must be equal to the value obtained when transforming both the field and the measurement device $|\langle d | X^\dagger X | f \rangle|^2$, because otherwise the measurement outcomes would depend on the reference frame. The non-Poincaré members of $C_{15}(3, 1)$ can be understood as changes of measurements units,^[67,68] which arguably makes the name “change of measurement frame” better than “change of reference frame” for $C_{15}(3, 1)$.

The conformal invariance of the scalar product also ensures that a single photon $\langle f | f \rangle = 1$, remains a single photon in all possible measurement frames. Such quantization is hence a conformally invariant concept.

Finally, it is important for completeness and future reference to include here two expressions for the scalar product that appear in the seminal papers of Gross and Zeldovich,^[69,70] in the coordinate (\mathbf{r}, t) and Fourier \mathbf{k} representations, respectively. Instead of using electric and magnetic fields, it is more convenient here to write them as a function of the helical combinations with $\lambda = \pm 1$:

$$\mathbf{F}_\lambda(\mathbf{r}, t) = \sqrt{\frac{\epsilon_0}{2}} [\mathbf{E}(\mathbf{r}, t) + i\lambda c_0 \mathbf{B}(\mathbf{r}, t)]$$

$$= \int_{\mathbb{R}^3 - \{0\}} \frac{d^3 \mathbf{k}}{\sqrt{(2\pi)^3}} \mathbf{F}_\pm(\mathbf{k}) \exp(i\mathbf{k} \cdot \mathbf{r} - i c_0 k t) \quad (21)$$

The $\mathbf{F}_\lambda(\mathbf{r}, t)$ are the positive frequency restriction of the Riemann–Silberstein vectors,^[71] and its monochromatic components are also known as Beltrami fields.^[85] As discussed above, the $\mathbf{F}_\lambda(\mathbf{r}, t)$ split any electromagnetic field into its left and right circular polarization handedness. The restriction to positive frequencies makes $\mathbf{E}(\mathbf{r}, t)$ and $\mathbf{B}(\mathbf{r}, t)$ necessarily complex valued, and is crucial for achieving the handedness splitting: If $\mathbf{E}(\mathbf{r}, t)$ and $\mathbf{B}(\mathbf{r}, t)$ are real-valued, then $|\mathbf{F}_+(\mathbf{r}, t)| = |\mathbf{F}_-(\mathbf{r}, t)|$ follows from the first line of Equation (21), which negates the handedness separation.

The expression of the scalar product used by Gross^[69] can be written as:

$$\langle f | g \rangle = \int_{\mathbb{R}^3 - \{0\}} \frac{d^3 k}{\hbar c_0 |\mathbf{k}|} \begin{bmatrix} \mathbf{F}_+(\mathbf{k}) \\ \mathbf{F}_-(\mathbf{k}) \end{bmatrix}^\dagger \begin{bmatrix} \mathbf{G}_+(\mathbf{k}) \\ \mathbf{G}_-(\mathbf{k}) \end{bmatrix} \quad (22)$$

and the one used by Zeldovich^[70] as:

$$2\pi^2 \hbar c_0 \langle f | g \rangle = \int_{\mathbb{R}^3} d\mathbf{r} \int_{\mathbb{R}^3} d\mathbf{r}' \frac{\mathbf{F}_+(\mathbf{r}, t)^\dagger \mathbf{G}_+(\mathbf{r}', t) + \mathbf{F}_-(\mathbf{r}, t)^\dagger \mathbf{G}_-(\mathbf{r}', t)}{|\mathbf{r} - \mathbf{r}'|^2} \quad (23)$$

4. The Computational Side

This section reviews two connections between the algebraic formalism and popular computational strategies in light–matter interaction: The T-matrix, and generic Maxwell solvers such as COMSOL, MEEP, JCMsuite, CST, or Lumerical.

4.1. The T-Matrix

The abstract Hilbert space formalism has a natural computational complement: The T-matrix. The T-matrix is a popular and powerful approach to the computation of light–matter interactions.^[86,88,137] The T-matrix encodes the full linear electromagnetic response of a given object upon arbitrary illuminations. The connection with the Hilbert space formalism is the simple bijective relationship that exist between the T-matrix and the scattering operator S :

$$S = I + T, \quad (24)$$

where I is the identity operator. The relationship is different from the typical $S = I + 2T$ of the monochromatic case, because of the conventions in Sections 3 and 3.1, which are more convenient for the polychromatic formalism. The T in Equation (24) should be called the T-operator for consistency, however, the name T-matrix is kept to avoid enlarging the terminology.

In computer calculations, the T-matrix is an actual matrix whose size and underlying basis are chosen with regard to robustness and convenience. For example, the multipolar basis is used in nanophotonics for describing molecules, microstructures, or small clusters thereof. The elements of the T-matrix are then:

$$T_{jm\lambda}^{\bar{j}\bar{m}\bar{\lambda}}(q, k) = \langle \bar{\lambda} \bar{m} \bar{j} q | T | k j m \lambda \rangle, \quad (25)$$

where the largest multipolar order j_{\max} in $j \in [1, 2, \dots, j_{\max}]$, and the range of the wavenumber $k(q) \in [k_{\min}, k_{\max}]$ must be chosen

so that the excluded terms do not affect the results significantly. Additionally, the $k(q)$ interval must be discretized with enough resolution. Equation (25) can be interpreted as follows: The T-matrix is applied to the multipolar state $|kj m \lambda\rangle$, and the resulting state is projected onto a different multipolar state, namely $|q \bar{j} \bar{m} \bar{\lambda}\rangle$.

In the multipolar basis, the result of $|g\rangle^{\text{out}} = S |f\rangle^{\text{in}}$, that is, the coefficients $g_{jm\lambda}(q)$ that determine $|g\rangle^{\text{out}}$, are computed as:

$$\begin{aligned} g_{jm\lambda}(q) &= \sum_{\lambda=\pm 1} \sum_{j=1}^{j_{\max}} \sum_{m=-j}^j \int_{k_{\min}}^{k_{\max}} dk k \underline{\underline{T}}_{jm\lambda}^{\bar{j}\bar{m}\bar{\lambda}}(q, k) f_{jm\lambda}(k) \\ &= f_{jm\lambda}(q) + \sum_{\lambda=\pm 1} \sum_{j=1}^{j_{\max}} \sum_{m=-j}^j \int_{k_{\min}}^{k_{\max}} dk k \underline{\underline{T}}_{jm\lambda}^{\bar{j}\bar{m}\bar{\lambda}}(q, k) f_{jm\lambda}(k) \end{aligned} \quad (26)$$

where the second equality follows from Equation (24), and the integrals can be implemented as Riemann sums over the discrete values of k and q .

In many cases, the matrices $\underline{\underline{T}}_{jm\lambda}^{\bar{j}\bar{m}\bar{\lambda}}(q, k)$ and $\underline{\underline{T}}_{jm\lambda}^{\bar{j}\bar{m}\bar{\lambda}}(q, k)$ are diagonal in frequency. Then, they can be obtained from the typical monochromatic T-matrices computed across the desired spectral range. Such explicit connection [10, Sec. 3.1-3.2] allows one to implement the polychromatic setting using the vast literature^[88,137] and the many public resources for monochromatic T-matrices.^[100,101] The formulas in Ref. [10, Sec. 3.1-3.2] assume that the monochromatic T-matrices are computed with the conventions in `treams`.^[99]

The plane wave basis is preferred for extended objects such as periodic metasurfaces. Then:

$$\underline{\underline{T}}_{\bar{\lambda}}^{\bar{\lambda}}(\mathbf{q}, \mathbf{k}) = \langle \bar{\lambda} \mathbf{q} | T | \mathbf{k} \lambda \rangle, \quad (27)$$

and the discretization of $\mathbf{k}(q)$ in momentum space must be chosen carefully, particularly in diffracting systems. Section 7 contains a brief discussion about polychromatic basis vectors defined by four discrete indexes, which would simplify the formalism.

There are many publicly available T-matrix codes^[100,101] that use Maxwell solvers to obtain T-matrices of objects such as dielectric disks and metallic helices. In the case of molecules, T-matrices in the dipolar approximation ($j_{\max} = 1$) can be obtained using quantum chemical simulations.^[93]

The following discussion illustrates the power and convenience of the combination of the algebraic formalism and the T-matrix. Let us consider the difference between the momentum of the fields along the z axis, before and after the interaction in Figure 1, which can be readily written down using scalar products [5]:

$$\begin{aligned} \langle \Delta P_z \rangle &= {}^{\text{in}}\langle f | P_z | f \rangle^{\text{in}} - {}^{\text{out}}\langle g | P_z | g \rangle^{\text{out}} \\ &= {}^{\text{in}}\langle f | P_z - S^\dagger P_z S | f \rangle^{\text{in}} \\ &= {}^{\text{in}}\langle f | -P_z T - T^\dagger P_z - T^\dagger P_z T | f \rangle^{\text{in}} \end{aligned} \quad (28)$$

The second and third equalities follow from Equation (1) and Equation (24), respectively. The last two expressions represent the momentum lost or gained by the object, computed as a function of the scattering operator and the T-matrix, respectively.

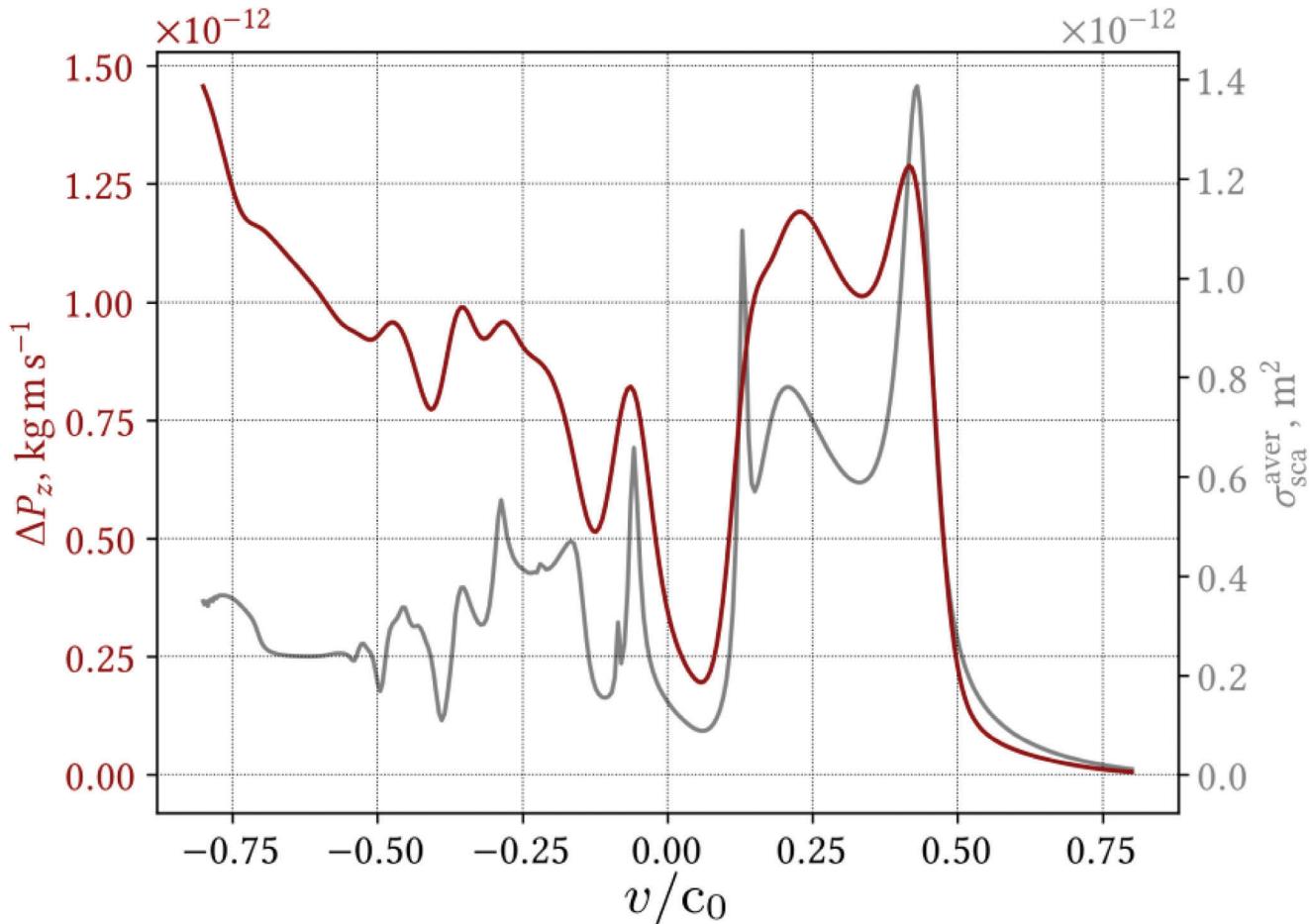


Figure 4. Adapted from Ref. [121]. The red line shows the transfer of momentum P_z from a light pulse to a silicon sphere moving with constant speed v along the z direction, either away or towards the light source, where positive v denotes movement away from the source. The gray line is the scattering cross-section. The results pertain to the reference frame of the sphere.

As with many other fundamental quantities, the change $\langle \Delta P_z \rangle$ of the momentum in the field is transferred to the material object. That is, Equation (28) is a way to compute the z -component of the optical force experienced by the object. Equations similar to Equation (28), albeit for the monochromatic case, are an important application of the T-matrix in optical traps and optical tweezers.^[12,13] While the results are identical to flux integrals of the Maxwell stress tensor and tensors derived from it, the scalar product expressions are arguably comparatively simpler. Once the T-matrix of a given object is known, the optical forces and torques exerted by any polychromatic field onto the object can be computed without solving Maxwell equations anymore.

The polychromatic setting allows one to treat similarly the case where the object is moving at relativistic speeds away or toward the light source with speed $v = \pm \hat{v}$, where the $+$ signs corresponds to movement away from the light source. For the transfer of momentum in the frame of reference of the object, Equation (28) is unchanged, however the incoming field is now a boosted version of the field $|f_0\rangle^{\text{in}}$ emitted by the source $|f\rangle^{\text{in}} = L_z(v)|f_0\rangle^{\text{in}}$, where $\hat{v} = \hat{z}$ is assumed without loss of generality. The calculations can be readily carried out.^[121] Figure 4 is an example thereof.

Expressions such as Equation (28) can also be used for optimization purposes. For example, to find out the most energy efficient polychromatic state for transferring angular momentum to a given object, that is, maximizing the optical torque per unit of energy:

$$|f\rangle^{\text{in}} \text{ that maximizes } \frac{\text{in}\langle f|J_z - S^\dagger J_z S|f\rangle^{\text{in}}}{\text{in}\langle f|H|f\rangle^{\text{in}}}, \quad (29)$$

whose solution is the generalized eigenstate of $J_z - S^\dagger J_z S$ and H with the largest generalized eigenvalue (see e.g., Ref. [138]). Generalized eigenstates are tightly related to the optical eigenstates introduced by Mazilu,^[14,15] which also facilitate optimization.

Similarly, one can obtain the most energy efficient pulse for maximizing the photon absorption, which could be useful in e.g., spectroscopy:

$$|f\rangle^{\text{in}} \text{ that maximizes } \frac{\text{in}\langle f|I - S^\dagger S|f\rangle^{\text{in}}}{\text{in}\langle f|H|f\rangle^{\text{in}}} \quad (30)$$

Such kind of optimizations are done in a computer over the finite set of coefficients $f_{jm\lambda}(k)$, or $f_\lambda(k)$. The generalized eigenvalue

decomposition problem is solved in several software packages, such as octave, matlab, and python (scipy.linalg).

4.2. The Electromagnetic Scalar Product in Spatially-Bounded Domains

This section explains how scalar products between pairs of outgoing or incoming fields can be computed when the fields are known only in spatially-bounded domains.

Quite often, generic Maxwell solvers are used for the study and optimization of photonic systems. Typically, a given photonic system is excited by a specified illumination, and the electromagnetic fields resulting from the light-matter interaction are obtained numerically. The benefits of the scalar product in this setting are manifold. For example, the knowledge of the number of photons of a given field in a simulation can be used to re-scale such field so that it contains one photon. Single-photon fields are needed in quantum nanophotonics for modeling single photon emitters, or for the photonic modes of objects such as nanoparticles or cavities.^[17] In another example, the total number of photons and the helicity of the field radiated by an emitter nearby a (chiral) nanostructure is a quantity of interest in the study of luminescence enhancement, in particular in chiral luminescence enhancement.^[139] However, none of the expressions of the scalar product that have appeared so far in here, namely Equations (11, 15, 22, 23), are directly applicable when the fields are known only in a spatially-bounded domain, which is the information available when using for example COMSOL, MEEP, JCMsuite, CST, or Lumerical. It is worth highlighting that laser interactions are very often studied within focal point interaction volumes. This problem is solved in Ref. [11] for incoming and outgoing fields by a new expression of the scalar product that only involves the values of the fields on any piecewise smooth surface ∂D enclosing a compact volume D containing the object, and is hence directly applicable to fields computed in a spatially-bounded domain (see Figure 5). The expression of the scalar product reads:

$$\langle f | g \rangle = \sum_{\lambda=\pm 1} (-\tau) i \lambda \int_{>0}^{\infty} \frac{dk}{\hbar c_0 k} \int_{y \in \partial D} d\mathbf{S}(y) \cdot [\mathbf{F}_\lambda^*(y, k) \times \mathbf{G}_\lambda(y, k)] \quad (31)$$

where $\tau = 1$ for outgoing fields, $\tau = -1$ for incoming fields, $d\mathbf{S}(y)$ is the unit vector perpendicular to the surface at point $y \in \partial D$, and Fourier transforms of the fields on the surface are used. For example:

$$\mathbf{F}_\lambda(y, k) = \int_{-\infty}^{\infty} \frac{dt}{\sqrt{2\pi}} \mathbf{F}_\lambda(y, t) \exp(i c_0 k t) \quad (32)$$

In single-frequency simulations, fields such as $\mathbf{F}_\lambda(y, k)$ are obtained directly. In time-domain simulations they can be obtained as integrals such as Equation (32), albeit with finite time integration limits because the fields on the surface will only be non-zero during a bounded period of time.

For outgoing(incoming) fields, the integral can be performed over any piecewise smooth surface ∂D enclosing a compact volume containing(excluding) the sources of radiation.

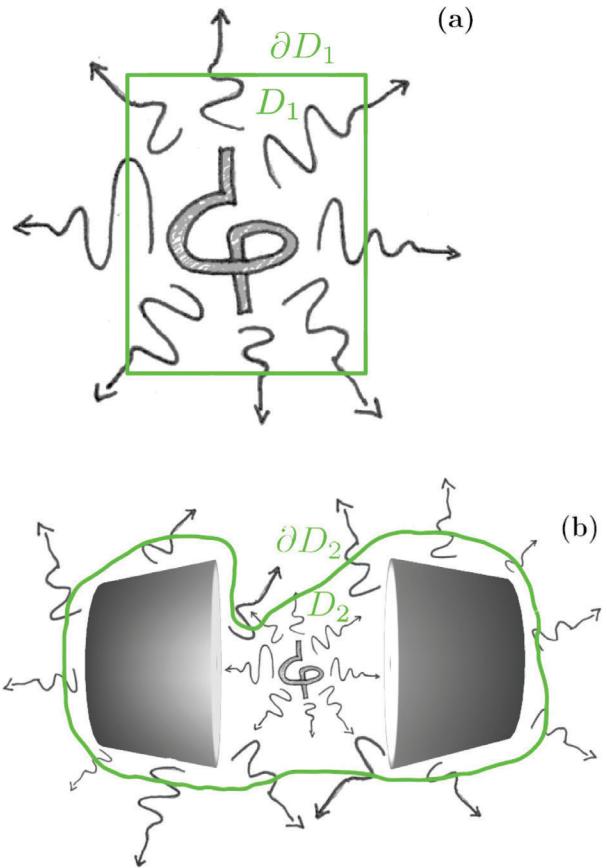


Figure 5. Figure from Ref. [11] Equations (33, 34, 35) can be used to compute the number of photons, energy, and helicity of the pulse emitted by the object in a first simulation (a), by means of integrals over the surface ∂D_1 . Then, the field can be scaled so that it contains a single photon, and used in a second simulation (b) as the emission that interacts with other objects. Integrals on the ∂D_2 surface provide the same quantities for the total outgoing field.

Using Equation (31), the number of photons, helicity, and energy of a given outgoing ($\tau = 1$) or incoming ($\tau = -1$) field can be computed as:

Number of photons $\langle f | f \rangle$

$$= \sum_{\lambda=\pm 1} (-\tau) i \lambda \int_{>0}^{\infty} \frac{dk}{\hbar c_0 k} \int_{y \in \partial D} d\mathbf{S}(y) \cdot [\mathbf{F}_\lambda^*(y, k) \times \mathbf{F}_\lambda(y, k)], \quad (33)$$

helicity $\langle f | \Lambda | f \rangle$

$$= \sum_{\lambda=\pm 1} (-\tau) i \int_{>0}^{\infty} \frac{dk}{c_0 k} \int_{y \in \partial D} d\mathbf{S}(y) \cdot [\mathbf{F}_\lambda^*(y, k) \times \mathbf{F}_\lambda(y, k)], \quad (34)$$

and energy $\langle f | H | f \rangle$

$$= \sum_{\lambda=\pm 1} (-\tau) i \lambda \int_{>0}^{\infty} dk \int_{y \in \partial D} d\mathbf{S}(y) \cdot [\mathbf{F}_\lambda^*(y, k) \times \mathbf{F}_\lambda(y, k)] \quad (35)$$

Similar formulas can be obtained for the change in the number of photons, energy, and helicity of the field upon light–matter interaction in scattering simulations [11, Eqs.(30,31,32)].

Besides augmenting the capabilities of the aforementioned numerical codes, which feature computations in spatial volumes, and are based on finite-difference time-domain, finite-element, or finite-difference frequency-domain methods, the computation of scalar products using the fields on spatial boundaries^[11] is perfectly adapted for boundary element methods.^[140,141] Similar formulas for the energy and helicity have been published very recently.^[142]

5. Symmetries, Selection Rules, and Quantitative Measures of Broken Symmetry

5.1. Symmetry in the Hilbert Space

Symmetry is one of the most general and useful concepts in physics. The symmetries of a physical system impose constraints, such as selection rules and conservation laws.^[143] When an effect observed in a particular system can be explained only by symmetry arguments, such as “the effect happens when the system has symmetry X and lacks symmetry Y,” the explanation is then valid and predictive in general: Systems that are vastly different from the original one will also exhibit the effect if they meet the symmetry requirements.

The algebraic setting for light–matter interactions allows one to treat symmetries and their consequences in a convenient and effective manner. Symmetry transformations are represented by unitary operators that act on the states in \mathbb{M} , mapping them back to \mathbb{M} . There are two kinds of symmetries, continuous such as translations, rotations, and electromagnetic duality, or discrete, such as parity or time reversal. Continuous symmetries depend on a real continuous parameter, called θ below, and are generated by exponentiating a self-adjoint operator, which ensures that the resulting symmetry operator is unitary:

$$X(\theta) = \exp\left(-i\frac{\theta}{\hbar}\Gamma\right) = \sum_{l=0}^{\infty} \frac{\left(-i\frac{\theta}{\hbar}\Gamma\right)^l}{l!},$$

with $\Gamma^\dagger = \Gamma$, then $X^\dagger(\theta) = X^{-1}(\theta)$ (36)

Some self-adjoint generators represent fundamental quantities such as energy, linear momentum, angular momentum and helicity. For example, the angular momentum operator J_z generates rotations along the z axis, $R_z(\theta) = \exp(-i\frac{\theta}{\hbar}J_z)$, and helicity Λ generates the electromagnetic duality transformation $D(\theta) = \exp(-i\frac{\theta}{\hbar}\Lambda)$, whose action on the fields is:

$$\begin{aligned} D(\theta)\mathbf{F}_+(\mathbf{r}, t) &= \exp(-i\theta)\mathbf{F}_+(\mathbf{r}, t), \\ D(\theta)\mathbf{F}_-(\mathbf{r}, t) &= \exp(i\theta)\mathbf{F}_-(\mathbf{r}, t), \\ D(\theta)\mathbf{E}(\mathbf{r}, t) &= \mathbf{E}(\mathbf{r}, t)\cos\theta - c_0\mathbf{B}(\mathbf{r}, t)\sin\theta, \\ D(\theta)c_0\mathbf{B}(\mathbf{r}, t) &= \mathbf{E}(\mathbf{r}, t)\sin\theta + c_0\mathbf{B}(\mathbf{r}, t)\cos\theta \end{aligned} \quad (37)$$

It is well-known that, besides $C_{15}(3, 1)$, Maxwell equation *without* sources are also invariant under the duality transformation,

which implies that the optical helicity in Equation (54) is a constant of the motion of free fields.

The action of a given continuous symmetry is particularly simple on eigenstates of its generator, for example:

$$R_z(\theta)|kjm\lambda\rangle = \exp(-im\theta)|kjm\lambda\rangle. \quad (38)$$

When an object is symmetric under a given unitary operator X , it means that the object is identical after transformation by X , which implies the following for its scattering operator S :

$$XSX^{-1} = S \iff [S, X] = 0 \iff [X, \Gamma] = 0, \quad (39)$$

where the first equivalence follows from the unitary character of X and the second, which only applies to continuous symmetries, can be readily shown by using the first line of Equation (36).

5.2. Selection Rules

Equation (39) leads immediately to the main consequence of symmetry in the algebraic setting: *Light–matter interaction with a symmetric object does not couple eigenstates of the symmetry with different eigenvalue*. This can be seen assuming that the incoming field is an eigenstate of X , $X|\gamma\rangle = \gamma|\gamma\rangle$, and projecting the outgoing field onto another eigenstate $X|\bar{\gamma}\rangle = \bar{\gamma}|\bar{\gamma}\rangle$:

$$\begin{aligned} \langle\bar{\gamma}|S|\gamma\rangle &= \langle\bar{\gamma}|XSX^{-1}|\gamma\rangle = (X^\dagger|\bar{\gamma}\rangle)^\dagger S X^{-1}|\gamma\rangle \\ &\Rightarrow (X^\dagger|\bar{\gamma}\rangle)^\dagger S(X^\dagger|\gamma\rangle) = \bar{\gamma}\gamma^* \langle\bar{\gamma}|S|\gamma\rangle, \end{aligned} \quad (40)$$

where the first equality follows from Equation (39), and the implication follows from the unitary condition of the symmetry operator: $X^\dagger = X^{-1}$. Equation (40) implies that $\langle\bar{\gamma}|S|\gamma\rangle = 0$ unless $\bar{\gamma}\gamma^* = 1$. Since $|\bar{\gamma}| = |\gamma| = 1$ because X is unitary, it follows that only $\gamma = \bar{\gamma}$ avoids the otherwise forceful zero.

The statement in italics above and Equation (40) is the precise formulation of what is commonly known as selection rules. Selection rules are extremely useful to understand what can and cannot happen in light–matter interactions *a priori*. Selection rules apply much independently of other aspects of the systems in question, such as for example whether far-field or near field effects are considered. For example, discrete rotational symmetries and helicity preservation upon on-axis illumination are sufficient conditions for ensuring zero backscattering in the far field,^[144] and so is a combined simultaneous discrete rotational-duality symmetry.^[145] Such conditions guide the design of anti-reflection coatings.^[146] A near-field example follows.

An emitter on top of a waveguide, as depicted in Figure 6, produces an electric ($\tau = 1$) or magnetic ($\tau = -1$) multipolar emission with well defined j and m [see Equation (7)]:

$$|f\rangle = \int_{>0}^{\infty} dk k f_{jm\tau}(k) |kjm\tau\rangle. \quad (41)$$

Since the waveguide is symmetric upon the mirror reflection M_z , $z \rightarrow -z$, its modes can be classified according to the eigenvalues of such symmetry, which are $\eta = 1$ for transverse magnetic (TM) modes, and $\eta = -1$ for transverse electric (TE) modes. When the

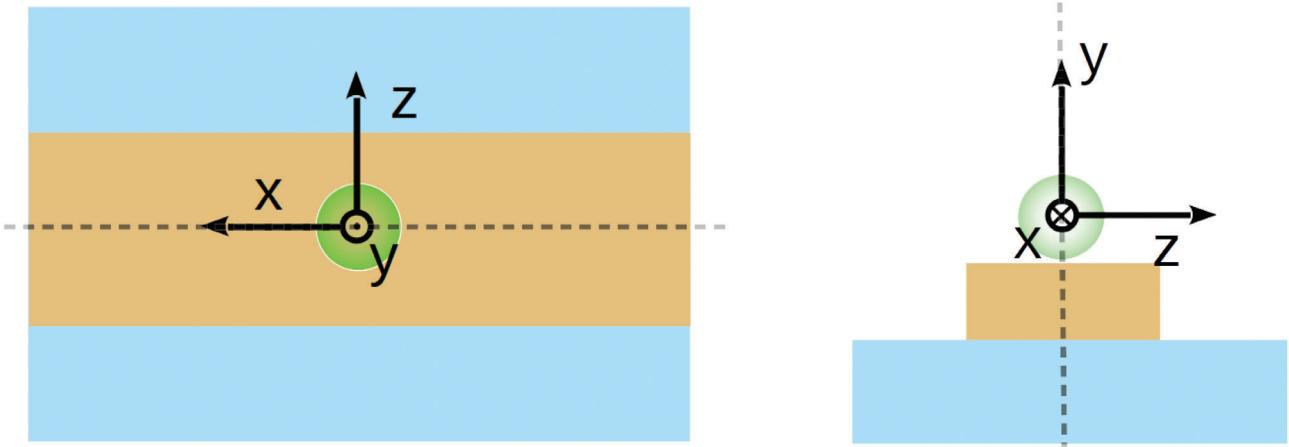


Figure 6. Selection rules can provide valuable a priori information with minimal effort. A waveguide (brown) on top of a substrate (blue) is invariant upon reflection $z \rightarrow -z$ across the XY plane, denoted by M_z . An electric or magnetic emitter (green) that is placed on the plane of symmetry on top of the waveguide will couple into the waveguide only when the M_z eigenvalues of the emission and the waveguide mode coincide. This results in the coupling selection rules for electric and magnetic dipolar and quadrupolar emissions onto TE or TM modes shown in Table 1.

emitter is placed on the plane of symmetry, its emission is also an eigenstate of M_z :

$$M_z |f\rangle = \int_{>0}^{\infty} dk k f_{jm\tau}(k) M_z |kjmt\rangle = \tau(-1)^{j+m} |f\rangle, \quad (42)$$

where the third equality follows from

$$\begin{aligned} M_z |kjmt\rangle &= \Pi R_z(\pi) |kjmt\rangle = (-1)^m \Pi |kjmt\rangle \\ &= \tau(-1)^{j+m} |kjmt\rangle, \end{aligned} \quad (43)$$

where Π is the parity operator. The last equality follows from Ref. [121, App. D.1.]

Then, the selection rule obtained from the M_z symmetry is:

$$\eta = \tau(-1)^{j+m}, \quad (44)$$

which predicts which electric or magnetic multipolar emissions can couple to which kind of waveguide modes, as seen in Table 1.

Table 1 is consistent with the “surface selection rule” where an emitter on an extended metal surface will have electric-dipole transitions quenched for transition moments parallel to the surface, but amplified for those perpendicular to it. According to the Table, the $m = 0$ parallel dipole component (along the z direction) can only couple to TE modes. However, the surface plasmons are TM modes. In contrast, The TM modes allow coupling to the $m = \pm 1$ electric dipoles, which contain y components perpendicular to the surface.

Table 1. Each combination of multipolar order j , angular momentum m , and electric/magnetic character of the emission can only couple to either a TE mode or a TM mode of the waveguide.

	$j = 1$				$j = 2$			
m	-1	0	1	-2	-1	0	1	2
(electric) $\tau = 1$	TM	TE	TM	TM	TE	TM	TE	TM
(magnetic) $\tau = -1$	TE	TM	TE	TE	TM	TE	TM	TE

It is remarkable that such detailed information can be obtained so easily, and without any numerical simulation. Similar considerations^[147] reveal the intricacies of the directional coupling of emitters onto waveguides.^[148–155] Understanding the origin of the selection rule also reveals ways for avoiding it by breaking the responsible symmetry. In the case of the system depicted in Figure 6, one can displace the emitter out of the plane of symmetry, or use a non-mirror symmetric waveguide.^[149]

5.3. Measures of Symmetry-Breaking

Symmetry-breaking is also an important concept in physics, which is relevant in phase transitions, chirality, and the Higgs boson, for example. While broken symmetries are often only qualitatively considered, the question *How much does the system break the symmetry?* can be quantitatively answered. Different symmetry-breaking measures have been defined for understanding spectra of nuclei and atoms,^[156,157] and for searching for hidden symmetries in the apparent disorder of liquids and colloidal glasses.^[21,22] Symmetry breaking measures are being studied in a more general sense,^[23–28,158] which has shown their usefulness in quantifying quantum resources, studying quantum state evolution and estimation, analyzing accidental degeneracies, and quantifying spontaneous symmetry breaking. Due to the generality of symmetry considerations, it is reasonable to expect many more areas of application, including currently unforeseen ones.

There is a rather straightforward way to measure symmetry breaking in the algebraic setting, which arises from the condition in Equation (39) that a symmetry operator X , and the scattering operator of a symmetric object meet: $S = XSX^{-1}$. An expression for the quantitative measurement of the breaking of any given symmetry X by any given object with scattering operator S was given in Ref. [6]:

$$m(S, X) = \frac{\|S - XSX^{-1}\|^2}{4\|S\|^2} \in [0, 1], \quad (45)$$

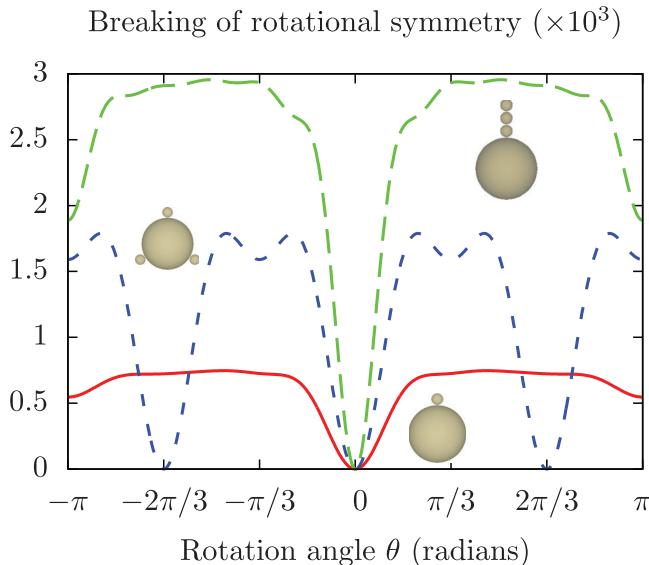


Figure 7. Figure from [6]. Dimensionless measure of the breaking of rotational symmetry by each of the three systems displayed inside the figure as a function of the rotation angle θ . The measure is computed using Equation (45), particularized to rotations with $X = R_z(\theta)$, and for a single frequency. In units of the wavelength, the big spheres have a radius of 0.5, the small spheres a radius of 0.1, and the minimal separation between spheres is 0.01. A relative permittivity equal to 10 was assumed for the material of all spheres. The fact that the green long-dashed line is always above the continuous red line agrees with the intuition that the system with three stacked small spheres should be more asymmetric than the system with one small sphere. The zeros at $\theta = \pm 2\pi/3$ of the blue short-dashed line reflect the discrete rotational symmetry of the corresponding system.

whose value is zero when the object is symmetric, and is upper bounded by 1, which denotes the maximum possible breaking of the symmetry. For a particular choice of the operator norm $\|A\|$, the quantity in Equation (45) can be obtained from experimental measurements.

Figure 7 shows the breaking of rotational symmetry that small spheres cause when placed around a bigger sphere. The calculations are, however, for a single frequency, since the polychromatic generalization was not available at the time. For the case of rotations by an angle θ , the denominator in Equation (45), $\|S - R_z(\theta)SR_z^{-1}(\theta)\|^2$, is a squared distance between the original scattering operator S , and the rotated version of itself $R_z(\theta)SR_z^{-1}(\theta)$. For the case of a cylindrically symmetric system, such distance is zero for all θ , otherwise, it measures how different are the original and rotated operators.

5.3.1. Electromagnetic Chirality

A discussion about symmetry breaking without addressing chirality misses an important aspect. Arguably, chirality is the most pervasive symmetry breaking in physics: From the left-right asymmetry of the weak interaction in particle physics, through the different response of chiral molecules to the two polarization handedness of light, to chiral magnetic fields of galactic scale. For an object to be chiral, it must lack parity symmetry, all mirror symmetries, and all rotation-reflection symmetries.^[159] While such definition is simple and intuitive, it is binary, and questions

of how chiral is a given object, or how to assign a handedness to it are much more difficult to answer than it may initially appear. The problems that such questions pose have been studied for many decades,^[29–36] and are clearly summarized by Fowler in Ref. [160]. For example, it is remarkable that, despite their broad use, the left and right-handed labels can not be appropriately assigned to all chiral objects.^[33,34] Such questions can, however, be answered in the context of light-matter interactions by scalar,^[45] and multidimensional measures^[122] of the electromagnetic chirality of an object. Such measures are continuous upon continuous changes of the object in question, avoid false chiral zeros and unhandled chiral states, and allow one to continuously distinguish any pair of enantiomers. The electromagnetic chirality measures are derived from the T-matrix of an object and, importantly, enjoy the highest possible degree of invariance in electromagnetism: Conformal invariance.^[122]

Both the scalar and the multidimensional electromagnetic chirality measures are obtained from the singular values of the sub-operators $T^{\lambda\bar{\lambda}}$, that is, the four restrictions of the T-matrix that map states of helicity $\bar{\lambda}$ to states of helicity λ . The ability to use powerful tools from the spectral theory of operators, such as the singular value decomposition, is a benefit of the algebraic approach.

6. Extension to Matter

The formalism outlined so far covers the “light-” part of the light-matter interaction, which motivates the following question: *Can this algebraic approach be used for the material object?* In this section, we review how it is possible to obtain expressions for fundamental quantities in matter using scalar products. This requires an appropriate mathematical representation of matter, an appropriate group of transformations, and a scalar product that is invariant under all the transformations of such group.

The representation of matter can be obtained by considering a version of Maxwell equations with sources [161, Sec. 5]:

$$\nabla \cdot \mathbf{B}(\mathbf{r}, t) = 0, \quad \nabla \times \mathbf{E}(\mathbf{r}, t) + \partial_t \mathbf{B}(\mathbf{r}) = 0, \quad \text{and}$$

$$\nabla \cdot \mathbf{E}(\mathbf{r}, t) = \frac{\rho(\mathbf{r}, t) - \nabla \cdot \mathbf{P}(\mathbf{r}, t)}{\epsilon_0}, \quad (46)$$

$$c_0^{-2} \nabla \times \mathbf{B}(\mathbf{r}, t) - \partial_t \mathbf{E}(\mathbf{r}, t) = \frac{1}{\epsilon_0} [\mathbf{J}(\mathbf{r}, t) + \partial_t \mathbf{P}(\mathbf{r}, t) + \nabla \times \mathbf{M}(\mathbf{r}, t)]$$

where the sources are the electric current four-vector containing $\rho(\mathbf{r}, t)$ and $\mathbf{J}(\mathbf{r}, t)$, and an antisymmetric tensor $\Sigma(\mathbf{r}, t)$ which contains both magnetization density $\mathbf{M}(\mathbf{r}, t)$, and polarization density $\mathbf{P}(\mathbf{r}, t)$:

$$j(\mathbf{r}, t) = \begin{bmatrix} \rho(\mathbf{r}, t) \\ \mathbf{J}(\mathbf{r}, t) \end{bmatrix}, \quad \Sigma(\mathbf{r}, t) = \begin{bmatrix} 0 & -c_0 P_1(t, \mathbf{r}) & -c_0 P_2(t, \mathbf{r}) & -c_0 P_3(t, \mathbf{r}) \\ c_0 P_1(t, \mathbf{r}) & 0 & -M_3(t, \mathbf{r}) & M_2(t, \mathbf{r}) \\ c_0 P_2(t, \mathbf{r}) & M_3(t, \mathbf{r}) & 0 & -M_1(t, \mathbf{r}) \\ c_0 P_3(t, \mathbf{r}) & -M_2(t, \mathbf{r}) & M_1(t, \mathbf{r}) & 0 \end{bmatrix} \quad (47)$$

Before the interaction starts and some time after the interaction finishes in Figure 1, the object is in static equilibrium, where the

time derivatives of all macroscopic quantities vanish. The sources are then assumed to be:

$$j(\mathbf{r}) = \begin{bmatrix} \rho(\mathbf{r}) \\ \mathbf{0} \end{bmatrix}, \Sigma(\mathbf{r}) = \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & -M_3(\mathbf{r}) & M_2(\mathbf{r}) \\ 0 & M_3(\mathbf{r}) & 0 & -M_1(\mathbf{r}) \\ 0 & -M_2(\mathbf{r}) & M_1(\mathbf{r}) & 0 \end{bmatrix}, \quad (48)$$

that is: Static sources can have electric charge and magnetization densities, but not magnetic charge or electric polarization densities. This is consistent with the fact that matter seems to feature electric charge and magnetic spin, but not its counterparts. Static electric polarization observed in ferroelectric materials or polar molecules can still be described by an appropriate distribution of the charge density $\rho(\mathbf{r})$. The movement of $\rho(\mathbf{r})$ results in the dynamic $j(\mathbf{r}, t)$, and the movement of $\mathbf{M}(\mathbf{r})$ results in $\Sigma(\mathbf{r}, t)$. Both kinds of dynamic sources can eventually radiate through Equation (46). The densities $\mathbf{P}(\mathbf{r}, t)$ and $\mathbf{M}(\mathbf{r}, t)$ are considered in recent works on fundamental quantities in matter.^[8,9,58]

Therefore, matter in static equilibrium can be represented by its charge density $\rho(\mathbf{r})$ and its magnetization density $\mathbf{M}(\mathbf{r})$, which are assumed to be contained in a finite volume. The similarity to the case of light is increased by using the static field $\mathbf{E}(\mathbf{r})$ produced by $\rho(\mathbf{r})$ instead of $\rho(\mathbf{r})$. The two are bijectively connected through the electrostatic equations:

$$\nabla \cdot \mathbf{E}(\mathbf{r}) = \frac{\rho(\mathbf{r})}{\epsilon_0}, \nabla \times \mathbf{E}(\mathbf{r}) = \mathbf{0}, \quad (49)$$

obtained as the static limits of equations in (46) by setting all time derivatives to zero, and removing $\mathbf{P}(\mathbf{r}, t)$. Static matter can hence be represented by the static field $\mathbf{E}(\mathbf{r})$ produced by its charge density, together with the static magnetization density $\mathbf{M}(\mathbf{r})$ produced by its magnetic spin texture: $|\Phi_{\omega=0}\rangle \equiv \{\mathbf{E}(\mathbf{r}), \mathbf{M}(\mathbf{r})\}$.

The next step is to find an appropriate group of invariance. To such end, one starts from the group of invariance of the dynamic equations, the conformal group $C_{15}(3, 1)$, and removes all the transformations that do not preserve the static $\omega = 0$ condition. Namely, Lorentz boosts and the time component of the special conformal transformations four-vector. Since time is not relevant anymore in the static case, the time translations can also be removed because, while preserving the $\omega = 0$ condition, any time-translations will just degenerate into the identity operator for the static case. Ten kinds of transformations are left: Space scalings, three spatial rotations, three spatial translations, and three special conformal transformations. Importantly, the remaining transformations also form a group: The conformal group in three-dimensional Euclidean space [162, Chapter 24], denoted by $C_{10}(3)$. It is rather remarkable and not at all a priori expected that removing transformations from a group results in another group. But in this case, the static restriction of the conformal group in 3+1 spacetime dimensions results in the conformal group in three spatial dimensions. Accordingly, $C_{10}(3)$ is taken as the group of transformations relevant for the static case.

The next step is to find a scalar product for material states $|\Phi_{\omega=0}\rangle \equiv \{\mathbf{E}(\mathbf{r}), \mathbf{M}(\mathbf{r})\}$, invariant under such group. It is shown in Ref. [9] that the following expression meets the invariance condition:

$$\langle \Phi_{\omega=0}^1 | \Phi_{\omega=0}^2 \rangle = \int_{\mathbb{R}^3} \frac{d^3k}{\hbar c_0 |\mathbf{k}|} \left[\frac{\sqrt{\epsilon_0}}{\sqrt{\mu_0}} \mathbf{E}^1(\mathbf{k}) \right]^\dagger \left[\frac{\sqrt{\epsilon_0}}{\sqrt{\mu_0}} \mathbf{E}^2(\mathbf{k}) \right]. \quad (50)$$

The Fourier transforms of the fields are used in Equation (50):

$$\mathbf{M}(\mathbf{k}) = \int_V \frac{d^3r}{\sqrt{(2\pi)^3}} \mathbf{M}(\mathbf{r}) \exp(-i\mathbf{k} \cdot \mathbf{r}), \text{ and} \\ \mathbf{E}(\mathbf{k}) = \frac{-i\mathbf{k}}{\epsilon_0 |\mathbf{k}|} \rho(\mathbf{k}) = \frac{-i\mathbf{k}}{\epsilon_0 |\mathbf{k}|} \int_V \frac{d^3r}{\sqrt{(2\pi)^3}} \rho(\mathbf{r}) \exp(-i\mathbf{k} \cdot \mathbf{r}), \quad (51)$$

where V is the finite volume occupied by the sources, and the first equality in the second line of Equation (51) follows from Equation (49). We note that both $\mathbf{E}(\mathbf{r})$ and $\mathbf{M}(\mathbf{r})$ are real-valued.

Self-adjoint operators such as the generators of $C_{10}(3)$ can now be “sandwiched” to obtain their value for a given $|\Phi_{\omega=0}\rangle$. Consistent with the assumption of static matter, is the fact that the values of linear and angular momentum vanish [9]:

$$0 = \langle \Phi_{\omega=0} | \mathbf{P}_\alpha | \Phi_{\omega=0} \rangle = \langle \Phi_{\omega=0} | \mathbf{J}_\alpha | \Phi_{\omega=0} \rangle, \alpha \in \{x, y, z\} \quad (52)$$

However, the value of some functions of those same operators do not vanish. For example, assuming that $\mathbf{M}(\mathbf{r}) = \mathbf{0}$, and considering the operator $c_0 |\mathbf{P}|$ for a charge density, one obtains that:

$$\langle \Phi_{\omega=0} | c_0 |\mathbf{P}| | \Phi_{\omega=0} \rangle = \epsilon_0 \int d^3k |\mathbf{E}(\mathbf{k})|^2 = \epsilon_0 \int_V d^3r |\mathbf{E}(\mathbf{r})|^2, \quad (53)$$

which is the expression for the electrostatic energy. Note that $c_0 |\mathbf{P}|$ is also one possible way to write the energy operator for Maxwell fields.

Even though their frequency content is different, light and matter are similarly represented in the Fourier \mathbf{k} domain. All the information is contained in $\{\mathbf{E}(\mathbf{k}), \mathbf{B}(\mathbf{k})\}$ for Maxwell fields, and in $\{\mathbf{E}(\mathbf{k}), \mathbf{M}(\mathbf{k})\}$ for matter. Such similarity allows one to extend towards the matter side some operators defined for Maxwell fields. The helicity operator in Equation (6) is one example. Helicity is a particularly interesting example because it encodes the chirality of Maxwell fields, and $\langle f | \Lambda | f \rangle$ is the difference between the number of left- and right-handed photons of a given field $|f\rangle$. This quantity is known as the optical helicity, whose most common expression is [124,125]:

$$\frac{1}{2} \int_{\mathbb{R}^3} d\mathbf{r} \mathcal{B}(\mathbf{r}, t) \cdot \mathcal{A}(\mathbf{r}, t) - \mathcal{E}(\mathbf{r}, t) \cdot \mathcal{C}(\mathbf{r}, t) \quad (54)$$

where $\mathcal{E}(\mathbf{r}, t)$ [$\mathcal{C}(\mathbf{r}, t)$] and $\mathcal{B}(\mathbf{r}, t)$ [$\mathcal{A}(\mathbf{r}, t)$] are the real-valued electric and magnetic fields[potentials], respectively. The use of two potentials is a common strategy in this context.^[125,130] In particular, it allows one to obtain an integrand which is local in \mathbf{r} , contrary to the case when only the fields are used. The value of Equation (54) coincides^[7] with the “sandwich” $\langle f | \Gamma | f \rangle$, which can be computed as:

$$\langle f | \Gamma | f \rangle = \int_{\mathbb{R}^3 - \{0\}} \frac{d^3k}{c_0 |\mathbf{k}|} |\mathbf{F}_+(\mathbf{k})|^2 - |\mathbf{F}_-(\mathbf{k})|^2 \quad (55)$$

or with any of the other expressions for the scalar product.

6.1. Helicity in Static Matter

The question of whether helicity is well defined in matter has been debated in the literature.^[49,163] The algebraic setting provides an affirmative answer, and an important connection:^[8]

$$\begin{aligned}\langle \Phi_{\omega=0} | \Lambda | \Phi_{\omega=0} \rangle &= \int_{\mathbb{R}^3} \frac{d^3 k}{c_0 |k|} \mu_0 \mathbf{M}(k)^\dagger [\hat{i}k \times \mathbf{M}(k)] \\ &= \int_{\mathbb{R}^3} \frac{d^3 r}{2Z_0} \mathbf{B}(r) \cdot \mathbf{A}(r)\end{aligned}\quad (56)$$

where $ik \times$ is the form of the helicity operator Λ in the \mathbf{k} domain, $Z_0 = \sqrt{\mu_0/\epsilon_0}$ is the impedance of vacuum, $\mathbf{B}(r)$ is the magnetic field produced by the magnetization density, defined by $\nabla \cdot \mathbf{B}(r) = 0$ and $\nabla \times \mathbf{B}(r) = \mu_0 \nabla \times \mathbf{M}(r)$, and $\mathbf{A}(r)$ is a vector potential such that $\nabla \times \mathbf{A}(r) = \mathbf{B}(r)$. The last expression in Equation (56) is essentially the magnetic helicity,^[104–106] which is typically defined as:

$$\int d^3 r \mathbf{B}(r) \cdot \mathbf{A}(r). \quad (57)$$

The magnetic helicity is relevant in diverse areas of physics such as cosmology,^[107,108] solar physics,^[109] fusion physics,^[110,111] magneto-hydrodynamics,^[112–115] and condensed matter.^[116–120]

The helicity in static matter $\langle \Phi_{\omega=0} | \Lambda | \Phi_{\omega=0} \rangle$ can be seen as a measure of the degree of twistedness of the static magnetization. The reason why the $\mathbf{E}(r)$ field does not contribute in Equation (56) is that $ik \times \mathbf{E}(k) = \mathbf{0}$, because the static field produced by any charge density distribution has zero curl [Equation (49)]. That means that the charge density does not contribute to the storage of helicity in static matter.

In this context, it is important to understand that the transformation generated by helicity, electromagnetic duality $D(\theta) = \exp(-i\theta\Lambda)$, has different effects in the static and dynamic cases. Instead of mixing the dynamic electric and magnetic fields as in Equation (37), duality acts on the static fields as follows:

$$\begin{aligned}D(\theta) \mathbf{E}(k) &= \mathbf{E}(k), \\ D(\theta) \mathbf{M}(k) &= D(\theta) [\mathbf{M}_+(k) + \mathbf{M}_-(k) + \mathbf{M}_0(k)] \\ &= \exp(-i\theta) \mathbf{M}_+(k) + \exp(i\theta) \mathbf{M}_-(k) + \mathbf{M}_0(k),\end{aligned}\quad (58)$$

where $\sum_{\lambda \in \{-1, 0, 1\}} \mathbf{M}_\lambda(k)$ is the decomposition of $\mathbf{M}(k)$ into helicity eigenstates. As opposed to the dynamic case where λ only takes the values -1 and $+1$, the eigenvalue 0 is possible in the static case.

The different action of duality is important, because if Equation (37) is assumed for the static case, then the expression of the magnetic helicity is not invariant under the transformation because there are no electric terms in it, which leads to the exclusion of the magnetic helicity as the material counterpart of the optical helicity.^[163] When the correct transformation in Equation (58) is considered, it readily follows that $\langle \Phi_{\omega=0} | \Lambda | \Phi_{\omega=0} \rangle$ is invariant under it, since:

$$\begin{aligned}\mathbf{M}(k)^\dagger [\hat{i}k \times \mathbf{M}(k)] &= \mathbf{M}(k)^\dagger [\mathbf{M}_+(k) - \mathbf{M}_-(k)] \\ &= |\mathbf{M}_+(k)|^2 - |\mathbf{M}_-(k)|^2\end{aligned}\quad (59)$$

Table 2. Adapted from [9] Helicity Λ , and angular momentum squared J^2 stored in a Hopfion. An analytical approximation of the Hopfion in a chiral FeGe magnet of cylindrical shape^[164] was used in the calculations. The height of the cylinder is equal to the magnetic helical period $L=90\text{nm}$, and the diameter is equal to $3L$. A magnetization density saturation value of $M_s=384\text{kA/m}$ is assumed. The charge density $\rho(r)$, and hence its electric field, is assumed to be zero: $|\Phi_{\omega=0}\rangle \equiv \{\mathbf{E}(r) = \mathbf{0}, \mathbf{M}(r)\}$. While a $\rho(r) \neq 0$ can affect the value of J^2 , it does not affect the value of Λ .

$\langle \Phi_{\omega=0} \Lambda \Phi_{\omega=0} \rangle$	$\langle \Phi_{\omega=0} J^2 \Phi_{\omega=0} \rangle$
$-129.1\hbar$	$1.30 \times 10^3 \hbar^2$

which is obviously invariant under duality, as seen from the last line of Equation (58). The longitudinal component of the magnetization does not store helicity because $ik \times \mathbf{M}_0(k) = \mathbf{0}$.

Equation (50) provides a means to compute fundamental quantities in matter for analytically derived,^[164] numerically obtained,^[165] or experimentally measured three-dimensional charge^[166] and magnetization density distributions.^[167] For computer calculations, it is possible to use the same sets of basis vectors that appear in Section 3, except that they must be complemented by adding their longitudinal counterparts, in other words, basis vectors that are eigenstates of helicity with eigenvalue zero. For example, adding a third kind of plane waves in Equation (13) with $\hat{e}_0(k) = \mathbf{k}$.

In an example of application, the helicity and total angular momentum squared of a given magnetic Hopfion can be computed, and are shown in Table 2. A magnetic Hopfion is a chiral magnetization configuration^[164,168–171] that can appear in chiral magnetic systems. Its magnetization vector forms a complex, knotted structure. Let us now consider the possibility of switching the handedness of the Hopfion by shining light on it. The connection between the optical helicity of the dynamic field, and the static helicity of the magnetization density suggests a lower bound for the number of circularly polarized photons that would be needed for switching the Hopfion onto its mirror image of opposite helicity: $[129.1 \times 2] = 259$. Additionally, $-129.1\hbar$ would also bound the helicity that can be radiated by the Hopfion as it loses its chirality, for example by the action of a large magnetic bias aligning its magnetization density vector along the same direction at all points. It is however yet to be demonstrated by either experiments or dynamic simulations of Maxwell fields interacting with $\Sigma(r, t)$ that helicity can be transferred between fields and matter. Connected to this is the question of whether a conservation law for the sum of the optical and magnetic helicities exist, albeit possibly under some restrictions. This open question is one of the differences between the transfer of angular momentum and the transfer of helicity. It is well-known that the isotropy of space-time implies the existence of a global conservation law for angular momentum, and that the transfer of angular momentum from light to matter imparts torque onto the material object. In contrast, the transfer of helicity, changing the degree of twistedness of the static magnetization, is a new subject for study. Another difference is that, while non-magnetic materials are not be

able to permanently store static helicity coming from the light field, the lack of static magnetization does not prevent light from exerting torque onto a given object. Differences are to be expected because, at the fundamental level and despite the fact that their units coincide, helicity and angular momentum are two different quantities, connected to two different symmetries, namely duality and rotational symmetry, respectively.

7. Conclusion and Outlook

This review has summarized an algebraic approach to light-matter interactions that is theoretically powerful and computationally friendly. Theoretical expressions can be developed and manipulated conveniently thanks to the generality of the basis on which the approach rests, and a compact notation. The tight connections to popular computational tools allow one to readily perform numerical calculations. After briefly indicating potential applications of the current framework, the article finishes with some research directions related to challenges and extensions.

The current setting is particularly suited for the computation of optical forces and torques, which can benefit experiments featuring optical levitation and manipulation of nanoparticles and molecules, specially when pulsed lasers are used. The ability to systematically handle changes of inertial reference frame^[121] can be applied to the prediction and understanding of electromagnetic measurements in astrophysics, such as radiation from pulsars, or spectroscopic measurements taken by a satellite orbiting a planet. Classical and quantum nanophotonics simulations using common Maxwell solvers can benefit from the properties of incoming and outgoing polychromatic fields for modeling absorption and emission by matter, respectively. For example, the photon emission of a molecule or a quantum dot, which has a definite start in time, can be constructed as an outgoing polychromatic field containing exactly one photon.

7.1. Outlook

The formalism would become simpler if orthonormal basis whose basis vectors were polychromatic fields in \mathbb{M} would be available for practical use. One such basis can be defined by extending a scalar result into \mathbb{M} [9, App. A]. Any such inherently polychromatic basis would allow one to write expansions with four discrete indexes, for example:

$$|f\rangle = \sum_{\lambda=\pm 1} \sum_n \sum_{j=1}^{\infty} \sum_{m=-j}^j c_{njm\lambda} |njm\lambda\rangle, \text{ with} \\ \sum_{\lambda=\pm 1} \sum_n \sum_{j=1}^{\infty} \sum_{m=-j}^j |c_{njm\lambda}|^2 < \infty, \text{ and} \\ \langle \bar{\lambda} \bar{m} \bar{j} \bar{n} | njm\lambda \rangle = \delta_{\bar{n}n} \delta_{\bar{j}j} \delta_{\bar{m}m} \delta_{\bar{\lambda}\lambda}.$$
 (60)

These polychromatic basis would simplify the formalism by avoiding the use of monochromatic basis, whose members are outside of \mathbb{M} , and would replace the corresponding wavenumber integrals by discrete sums that are easier to implement. For example, the discretization of the wavenumber is specially complicated when treating Lorentz boosts.^[121] The questions regarding

discretization of the wavenumber would be replaced by questions of how many terms to take in the sum over n . From the mathematical point of view, Equation (60) would establish a clean isomorphism between \mathbb{M} and ℓ^2 , the Hilbert space of square-summable sequences, and give access to results demonstrated for ℓ^2 .

The computational side of the framework would benefit from the resolution of an infamously outstanding issue in the T-matrix formalism: The region of validity of the scattered near-field computed as the last term of Equation (26). On the one hand, the expansion in spherical waves ensures accuracy only up to the surface of the smallest circumscribing sphere that contains the object. Such boundary is often violated in systems of interest such as a quantum dot nearby a nano-antenna. As a consequence, the computation of the electromagnetic coupling between the two objects is compromised. On the other hand, there seems to be enough information in the T-matrix for computing the electromagnetic fields up to the surface of the object, as the existence of several different techniques shows.^[94,95,172–177] The existing solutions are based on rather involved theoretical and numerical strategies. One can argue that the problem has not been fully solved in general, and hope for a simpler solution that would avoid the inconvenience and computational costs of the convergence checks that must be performed, for example, when computing the T-matrix of crystals when one unit cell invades the circumscribing sphere of the next unit cell.

It seems possible to extend the algebraic approach to bipartite states of light,^[178] such as biphoton entangled states,^[179–182] or the states involved in non-linear effects^[183–187] such as second harmonic generation (SHG). The extension is based on the tensor product of two copies of the Hilbert space \mathbb{M} , $\mathbb{M}_2 = \mathbb{M} \otimes \mathbb{M}$, restricted by the bosonic permutation. The extension of theories from a Hilbert space onto the tensor product of two copies of such Hilbert space, which is sometimes called “the double copy”, has already been used in the study of photonic bipartite entanglement, formulations of gravity, and analogies between light and gravitation, in particular mapping bipartite states of light to gravitational waves.^[188–192] Benefits of the extension to bipartite states are, for example, the computation of fundamental properties using scalar products, and the easy treatment of symmetries and their consequences for such states.

It is interesting to investigate potential connections between the Hilbert space formalism and the formalism of quasi-normal modes (QNMs), where the natural damped resonances of realistic nanophotonic systems are used for the study and engineering of light-matter interactions.^[193–197] The approach connects with the physics of resonators, and software packages implementing the QNM formalism are available.^[193] In one research direction, the orthogonality, normalization, and completeness properties of the QNMs can be studied using the conformally invariant scalar product. Additionally, a connection between the T-matrix of an object and its QNMs would create synergies between both approaches. For example, the computation of T-matrices would become more efficient if the response of the object can be described to good approximation with only a few QNMs, at least in limited frequency ranges.

One can argue that the conformal group should receive more research attention, for example regarding a systematic consideration of the inequivalent irreducible representations of the

conformal groups $C_{15}(3, 1)$ and $C_{10}(3)$ for describing radiation and matter, respectively, and also regarding the decomposition of the product group $C_{15}(3, 1) \times C_{10}(3)$ into its irreducible representations. Such product group is a plausible structure for describing hybrid light-matter entities such as “dressed” material states,^[198] and polaritons.

Finally, the transfer of helicity between light and matter discussed in Section 6.1 is an intriguing possibility. Such possibility is, however, yet to be demonstrated in either experiments or dynamic simulations. The dynamic simulations should consider the interactions between the fields, the current four-vector, and the magnetization-polarization tensor, including self-interactions. The question of whether a conservation law for the sum of the optical and static helicities exist, albeit possibly under some restrictions, is related to the transfer of helicity between light and matter. The potential impact of these research questions reaches different areas of physics and technology. For example, micromagnetics, regarding the all optical switching of magnetization with circularly polarized radiation,^[117] fusion physics, where the injection of magnetic helicity is considered for controlling the plasma,^[110,111] and cosmology, where helical magnetic fields with galactic-scale coherent lengths are studied.^[107,108]

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Conflict of Interest

The authors declare no conflict of interest.

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