Dipole-quadrupole spherical tensors in resonant x-ray diffraction

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Abstract. Parity-breaking phenomena in condensed-matter physics, either associated or not to time-reversal-breaking effects, are nowadays considered as an important part of fundamental and applied research, especially in connection to multiferroic materials. Resonant x-ray diffraction is probably the most suited experimental technique to study multiferroic order parameters. In the present note, after some introduction on multipolar order parameters, I provide the polarization and wave-vector dependence of the spherical tensors involved in parity-breaking effects in resonant x-ray diffraction.

1. Introduction
Parity and time-reversal breaking phenomena in condensed matter have recently received a big deal of attention, especially in connection to the possible technological applications of multiferroic materials, eg, in the field of spintronics [1]. However, such materials are very attractive even from a fundamental point of view, because of the interplay between magnetic and electric degrees of freedom, whose microscopic modelization is still far from clear.

In parallel, the research field in resonant x-ray diffraction (RXD) has achieved tremendous progress in the last years. Nowadays RXD is rapidly becoming the crucial technique for investigating the subtleties of microscopic parity and time-reversal symmetry-breaking phenomena in such systems [2].

The possibility to single out magnetic and non-magnetic scattering components through the Bragg diffraction, and to study the polarization dependence of the photon diffracted beam as a function of scattering angles (azimuthal scans) has provided a great amount of experimental data, that in turn stimulated the theoretical interpretation. In particular, the interpretation of the RXD signal in terms of electromagnetic multipoles is, at present, one of the most fashionable ways to classify the experiments. In the light of this interpretation, RXD has been applied to the investigations of many complex materials, such as manganites, actinides, multiferroics and other transition metal oxides, in which different degrees of freedom coexist (orbital, charge, magnetic, dipolar and octupolar, etc.), in such a way as to disentangle the different contributions [2].

The next two sections of the paper are devoted, respectively, to a brief summary of the recent advances in the use of RXD to detect multipolar order parameters (Section 2), and to the detailed expression of the polarization and wave-vector dependence of the RXD signal in...
the dipole-quadrupole channel (Section 3). Such a dependence had been already implemented in 2003 by me and Yves Joly in Joly’s program fdmnes [3] and used several times since then [4, 5, 6].

2. Multipolar expansion in crystals and resonant x-ray diffraction.

The aim of this section is to briefly introduce how it is possible to detect electric and magnetic multipoles by means of resonant x-ray spectroscopies and how to write the one-to-one correspondance of each multipole with the measurable quantities at K edge RXD.

In general the multipole expansion of electric and magnetic fields generated by fixed charges and permanent currents is a widely used tool to characterize the electromagnetic state of a physical system[7]. For example, for a charge distribution \( \rho(\vec{x}) \) in a given external potential field \( \Phi(\vec{x}) \), whose electrostatic energy is \( W_E = \int \text{d}^3x \rho(\vec{x}) \Phi(\vec{x}) \), one can exploit the well-known expansion: \( W_E = q \Phi(0) - \vec{d} \cdot \vec{E} - \frac{1}{b} \sum_{ij} Q_{ij} \frac{\partial \Phi}{\partial x_j}(0) + \ldots \). Here \( 0 \) is some properly chosen origin, and \( \vec{E} = -\nabla \Phi \). Such an expansion shows how the external field couples with the various multipoles of the charge distribution: the potential with the charge \( q \), the electric field with the dipole \( \vec{d} \), the gradient of the field with the quadrupole \( Q_{ij} \equiv 3x_i x_j - r^2 \delta_{ij} \), and so on. A similar expansion holds also for the vector potential. In this case there is no monopole term, of course, and the magnetic energy is: \( W_M = \int \text{d}^3x \ \vec{J}(\vec{x}) \cdot \vec{A}(\vec{x}) \). However, due to the vector character of the current density \( \vec{J} \) and the potential \( \vec{A} \), it is often useful to decompose them, before exploiting the expansion, in a longitudinal (rotor-free) and a transversal (divergenceless) part: \( \vec{J} = \vec{J}_\parallel + \vec{J}_\perp \). As \( \nabla \cdot \vec{J} = \nabla \cdot \vec{J}_\parallel \equiv -\partial_t \rho \), then the longitudinal \( \vec{J}_\parallel \) is related to the time-derivative of the charge multipoles. On the other side, the solenoidal field \( \vec{J}_\perp \) is characterized by two independent families of moments, and can be written, in the Helmoltz-Debye representation, as: \( \vec{J}_\perp = \vec{l} \psi(\vec{x}) + \nabla \times (\vec{l} \chi(\vec{x})) \), where \( \vec{l} \) is the orbital angular momentum and \( \psi(\vec{x}) \) and \( \chi(\vec{x}) \) are a scalar and pseudoscalar function, respectively. The first vector describes the toroidal currents (flowing along the parallels on a sphere), and the second the poloidal currents (flowing along the meridians on a torus). Toroidal currents are widely used, eg, in the physics of electron plasma. Following Ref. [8], we define the magnetic multipoles of the two families in terms of the orbital angular momentum; when also spin quantities play a role, it is sufficient to consider the substitution \( \vec{l} \rightarrow \vec{l} + g_s \vec{s} \), where \( g_s \) is the spin giromagnetic ratio. Then we have:

- a) Magnetic moment: \( \vec{m} = \frac{e}{2m} \vec{l} = \mu_B \vec{l} \);
- b) Magnetic quadrupole: \( m_{ij} = \mu_B (x_i l_j + l_i x_j) \);
- c) Magnetic (polar) toroidal moment: \( \vec{t} = \frac{1}{2} \mu_B \vec{m} \times \vec{m} \); \( \vec{t} = \frac{1}{2} \mu_B \vec{m} \times \vec{m} \);
- d) Magnetic toroidal quadrupole: \( t_{ij} = x_i t_j + t_i x_j \).

Notice that the magnetic toroidal moment is known in high-energy physics as anapole[9]. A simple geometrical image of \( \vec{t} \) is a closed circle of elementary magnets \( \vec{m} \) joined to each other. By analogy with this image, one can define a similar quantity[10], by replacing \( \vec{m} \) with the electric dipole \( \vec{d} \). Then we get:

- e) Axial toroidal moment: \( \vec{g} = \frac{1}{2} (\vec{x} \times \vec{d}) \); \( \vec{g} = \frac{1}{2} (\vec{x} \times \vec{d}) \);
- f) Axial toroidal quadrupole: \( g_{ij} = x_i g_j + g_i x_j \).

These moments are scalarly coupled with the corresponding fields, as detailed, eg, in Ref. [8].

How can these order parameters be detected by means of RXD?

In order to answer this question, we ought to write down the equations used to describe RXD. Resonant x-ray diffraction (or resonant x-ray scattering as it is often termed) is a technique that takes advantage of the sensitivity of usual x-ray diffraction to ordered structure and of the sensitivity of electron resonances to local density of states. It is a core resonant spectroscopies that depends on the virtual processes that allow promoting a core electron to some empty energy levels. All these processes can be described by the transition matrix elements of matter-radiation interaction:
\[ M_{\text{ng}}^{i(o)}(j) = \langle \psi_n | \hat{O}^{i(o)} | \psi_g(j) \rangle \] (1)

where, in the x-ray regime, the operator \( \hat{O} \) is written through the multipolar expansion of the photon field up to electric dipole (E1) and quadrupole (E2) terms [11]:

\[ \hat{O}^{i(o)} = \varepsilon^{i(o)} \cdot \vec{r} (1 - \frac{1}{2} i \hat{k}^{i(o)} \cdot \vec{r}) \] (2)

In Eq. (1), \( \psi_g(j) \) is the core ground state centered around the \( j^{th} \) atom and \( \psi_n \) the probe photo-excited state, whereas in Eq. (2), \( \vec{r} \) is the electron position measured from the absorbing ion, \( \varepsilon^{i(o)} \) the polarization of the incoming (outgoing) photon and \( \hat{k}^{i(o)} \) its corresponding wave vector. In RXD the global process of photon absorption, virtual photoelectron excitation and photon re-emission, is coherent throughout the crystal, thus giving rise to the usual Bragg diffraction condition:

\[ F = \sum_j e^{i\vec{Q} \cdot \vec{R}_j} (f_0 + f'_j + if''_j) \] (3)

Here \( \vec{R}_j \) stands for the position of the scattering ion \( j \), \( \vec{Q} \) is the diffraction vector and \( f_0 \) is the usual Thomson factor. The resonant part, \( f'_j + if''_j \equiv f_j \), is the anomalous atomic scattering factor, given by the expression [12]:

\[ f_j(\omega) = \frac{m_e}{\hbar^2} \frac{1}{\hbar \omega} \sum_n \frac{(E_n - E_g)^3 M_{\text{ng}}^{\alpha}(j) M_{\text{ng}}^{i}(j)}{\hbar \omega - (E_n - E_g) - i \frac{\Gamma_n}{2}} \] (4)

where \( \hbar \omega \) is the photon energy, \( m_e \) the electron mass, \( E_g \) the ground state energy, and \( E_n \) and \( \Gamma_n \) are the energy and inverse lifetime of the excited states. The sum is extended over all the excited states of the system.

The matrix element in Eq. (1) depends only on the electronic part of the operator \( \hat{O} \), so that the radiation parameters \( \varepsilon \) and \( \hat{k} \) can be factorized out. What one gets is therefore:

\[ f_j(\omega) = \sum_{\alpha \beta} \epsilon^{\alpha \beta} \epsilon^{i \beta} F^{DD}_{\alpha \beta}(j; \omega) \]

\[ -\frac{i}{2} \sum_{\alpha \beta \gamma} \epsilon^{\alpha \beta} \epsilon^{i \beta} \left( k^{\alpha} F^{DQ}_{\alpha \beta \gamma}(j; \omega) - k^{\beta} F^{DQ}_{\alpha \beta \gamma}(j; \omega) \right) \]

\[ + \frac{1}{4} \sum_{\alpha \beta \gamma \delta} \epsilon^{\alpha \beta} \epsilon^{i \beta} k^{\alpha} k^{\beta} F^{QQ}_{\alpha \beta \gamma \delta}(j; \omega) \] (5)

where we have explicitly written the tensor dependence of the scattering amplitude. The terms \( F^{DD}_{\alpha \beta} \), \( F^{DQ(QD)}_{\alpha \beta \gamma} \), and \( F^{QQ}_{\alpha \beta \gamma \delta} \) represent all what is left of Eq. (4) once the polarization and wave vector components are extracted from the transition matrix elements in the dipole-dipole, dipole-quadrupole and quadrupole-quadrupole channels, respectively, whereas \( \alpha \beta \gamma \delta \) represent cartesian components \((x, y, z)\). The interest for such a separation can be easily explained as follows: today polarization detectors in the x-ray range allow a reliable polarization analysis of the diffracted signal. At the same time, the incoming x-rays can also be completely polarized. This allows analysing different cartesian components of the signal by varying polarization and wave-vector directions of incoming and outgoing beams, thereby exploring the intrinsic anisotropies of the charge distribution, what in the earlier literature on the subject [13, 14] was called anisotropic tensor of susceptibilities.
However, it is possible to provide a much deeper physical insight if we re-write Eq. (6) in terms of the scalar product of irreducible tensors \([15, 16, 4]\), instead of using cartesian tensors. Formally, one can then re-write Eq. (6) as \(f_j(\omega) = \sum_{p,q} (-)^q T_{q}^{(p)} F_{-q}^{(p)} (j; \omega)\). Here \(T_{q}^{(p)}\) depends only on the incident and scattered polarization and wave vectors, ie, it is the spherical counterpart of the polarization and wave-vector terms in Eq. (6), while \(F_{-q}^{(p)} (j; \omega)\) represents the properties of the system under study, ie, it is the spherical counterpart of the \(F^{DD}, F^{DQ(QD)}\) and \(F^{QQ}\) terms. The advantage obtained with this reformulation is that the rank \(p\) of these irreducible tensors represents the order of the multipoles in the electromagnetic field expansion (see, e.g., Refs. [4, 15, 17]). For example, for each \(p\), \(F_{q}^{(p)} (j; \omega)\) is related to a specific term of the multipolar expansion of the system, as shown in Ref. [4]. The allowed \(p\) are, in the dipole-dipole channel \(p = 0, 1, 2\), in the dipole-quadrupole channel \(p = 1, 2, 3\) and in the quadrupole-quadrupole channel \(p = 0, 1, 2, 3, 4\). As usual, \(q\) is an integer between \(-p\) and \(p\). For each \(p\), \(F_{q}^{(p)} (j; \omega)\) is related to a specific term of the multipolar expansion of the system, according to the following table:

| tensor | \(\hat{T}\) | \(\hat{P}\) | multipole |
|--------|-------------|-------------|-----------|
| \(F^{(0)}(E1 - E1)\) | + | + | electric charge |
| \(F^{(1)}(E1 - E1)\) | - | + | magnetic dipole |
| \(F^{(2)}(E1 - E1)\) | + | + | electric quadrupole |
| \(F^{(1+)}(E1 - E2)\) | + | - | electric dipole |
| \(F^{(2+)}(E1 - E2)\) | + | - | axialtoroidalquadrupole |
| \(F^{(3+)}(E1 - E2)\) | + | + | electric octupole |
| \(F^{(1-)}(E1 - E2)\) | - | - | polar toroidal dipole |
| \(F^{(2-)}(E1 - E2)\) | - | + | magnetic quadrupole |
| \(F^{(3-)}(E1 - E2)\) | - | + | polar toroidal octupole |
| \(F^{(4)}(E2 - E2)\) | + | + | electric hexadecapole |

For \(p = 0, 1, 2\), E1–E1 and E2–E2 tensors represent the same physical quantities, though referred to states with different angular momentum. The identification illustrated in Table I is dictated by the unique properties under time-reversal (\(\hat{T}\)) and parity (\(\hat{P}\)) reflections of \(F^{(p)}\) tensors on one side and multipole terms on the other. In fact, for any given tensor rank \(p = 1, 2, 3, 4\), there is just one electromagnetic multipole of the same rank (1→dipole, 2→quadrupole, 3→octupole, 4→hexadecapole) with the same \(\hat{T}\) and \(\hat{P}\) properties. Notice that \(\hat{P}\)-odd E1–E2 tensors have both \(\hat{T}\)-odd (-) and \(\hat{T}\)-even (+) terms for any \(p\) ([17]), while \(\hat{P}\)-even tensors (both E1–E1 and E2–E2) are \(\hat{T}\)-odd if of odd rank and \(\hat{T}\)-even if of even rank.

As each \(F^{(p)}\) is scalarly related to a corresponding \(T^{(p)}\), they have the same \(\hat{T}\) and \(\hat{P}\) properties. For example, in the E1–E1 or E1–E2 channels, \(T^{(0)} \equiv \hat{e}_o \cdot \hat{e}_i\) is a scalar and \(T^{(1)} \equiv \hat{e}_o \times \hat{e}_i\) is a \(\hat{T}\)-odd and \(\hat{P}\)-even vector. Analogously, in the E1–E2 channel, the previous quantities combine with the two \(\hat{P}\)-odd vectors \(\hat{k}^3 + \hat{k}^o\), which is \(\hat{T}\)-odd, and \(\hat{k}^3 - \hat{k}^o\), which is \(\hat{T}\)-even.

A comment here is required: up to now, we have deliberately avoided the question of whether the local density of the tensors probed by these spectroscopy has anything to do with the properties of the ground state, which we are interested in. At first sight, the presence of the core hole in the final state would prone to give a negative answer to this question, and yet, the wide success of sum rules for magnetic circular dichroism[18] seems to prove the contrary. Moreover, in Ref. [17] in the framework of multiple scattering theory, it was demonstrated that these spectroscopies are sensitive to the expectation values of some tensor operators in the scattering
states at the energy of the measurement, so that, by integrating over a specific edge, sum rules can be recovered[19]. However, a definitive answer to this problem appears far from clear, as the introduction of the so-called 'breathing' parameters for 5d radial wave functions in the case of rare-earths L_{2,3}-edges [20] stands to indicate (see in particular Refs. [21, 22]). In the following we assume that we may refer to the values of the multipoles in the ground-state, but all these ‘warnings’ should always be present to the reader.

All the quantities described in Table I are detectable by means of RXD, where the local transition amplitudes are added with a phase factor that can compensate the possible vanishing effect due to the global symmetry. RXD allows us to gain information about the following set of electromagnetic multipoles (the first five are parity-even, E1–E1 or E2–E2, the latters parity-odd, E1–E2):

1) Charge, expressed by \( F(0) \). This is usually the strongest contribution. This term allows us to prove (or disprove) the so-called charge-ordering through forbidden diffraction-peaks analysis (see, e.g., Ref. [23]). In fact, any modification of the atomic charge implies a core level shift, and the measure of the energy shift of the threshold allows us to obtain the total valence charge with a good resolution.

2) Magnetic moments (ie, \( F(1)(E1 − E1) \)) have been treated in a widespread context, due to the big success of the "Carra-Thole-van der Laan" sum-rules[18]. In RXD, this term has been detected several times, for example in V\(_2\)O\(_3\) [24] and KCuF\(_3\) [25].

3) Electric quadrupole (\( F(2)(E1 − E1) \)) is the biggest non-magnetic anisotropic term, responsible, e.g., for structural linear dichroism, as well as, in RXD signals, for the anomalous Jahn-Teller and "orbital ordering" peaks in systems like manganites and KCuF\(_3\) [25]. It is often called Templeton scattering, from the name of its discoverer [13].

4) Magnetic octupole (\( F(3)(E2 − E2) \)) was invoked in Ref. [26] to explain the anomalous RXD signal in NpO\(_2\) and was proved to give a contribution in the anomalous magnetic signal in V\(_2\)O\(_3\) [24].

5) Electric hexadecapole (\( F(4)(E2 − E2) \)) was detected in \( \alpha \)-haematite by Finkelstein and collaborators, as explained in Ref. [15], and detected in Ref. [24].

6) Electric dipole (\( F(1+)(E1 − E2) \)) is observable by RXD in antiferroelectric materials, like ErGe\(_3\) or VOMoO\(_4\), as theoretically predicted in Ref. [27], or experimentally observed in K\(_2\)CrO\(_4\) [28].

7) Non-magnetic (axial) toroidal quadrupole was found with XNCD in \( \alpha \)-LiIO\(_3\),[29] and in V\(_2\)O\(_3\) by means of RXD[16].

8) Electric octupole (\( F(3+)(E1 − E2) \)) has been predicted in Ref. [30] for K\(_2\)CrO\(_4\) on the basis of symmetry arguments and detected in Ref. [28].

9) Magnetic toroidal moment (\( F(1−)(E1 − E2) \)) and octupole (\( F(3−)(E1 − E2) \)), together with magnetic quadrupole (\( F(2−)(E1 − E2) \)) have been found in the anomalous magnetic signal of V\(_2\)O\(_3\)[24, 31], and, more recently, in the magnetoferroelectric material GaFeO\(_3\) [32, 5].

3. Classification of dipole-quadrupole terms: polarization and wave-vector dependence.

This section is devoted to writing down the explicit expression for the polarization and wave-vector dependence of E1-E2 terms in RXD, in order to allow disentangling the different contributions by azimuthal scan in RXD. This is quite simple when the symmetry allows only a few multipoles (as shown, eg, in Ref. [16] for corundum systems at the (00.3)\( _h \) reflection where only the electric hexadecapole and axial toroidal quadrupole are present). It might however become very complicate for low-symmetry systems, due to the intrinsic experimental uncertainty in the relative intensity of an experimental azimuthal scan (ie, one should be sure that the sample rotation is ideal in such a way as to probe always the same volume in the sample).
Theoretically, however, the procedure is well defined, once we have the explicit dependence on geometrical conditions (polarization and wave-vector) of each single multipolar term, as given below.

If we write:

\[ F_{\alpha\beta\gamma}^{DQ}(j;\omega) = \frac{m_e}{\hbar^2\hbar\omega} \sum_n \frac{(E_n - E_g)^3 I_{\alpha\beta\gamma}(j, n)}{h\omega - (E_n - E_g) - i\frac{\gamma}{2}} \]  

(6)

and

\[ F_{\alpha\beta\gamma}^{QD}(j;\omega) = \frac{m_e}{\hbar^2\hbar\omega} \sum_n \frac{(E_n - E_g)^3 I_{\alpha\beta\gamma}(j, n)}{h\omega - (E_n - E_g) - i\frac{\gamma}{2}} \]  

(7)

where \( I_{\alpha\beta\gamma}(j, n) = \langle \psi_g(j) | r_\alpha | \psi_n(j) \rangle \langle \psi_n | r_\beta r_\gamma | \psi_g(j) \rangle \), then the dipole-quadrupole part of the scattering amplitude in Eq. 6 can be rewritten after some algebraic manipulations as:

\[ f_j^{DQ}(\omega) = \frac{i}{2} \sum_n \text{Den}(n, \omega, \Gamma) \sum_{\alpha\beta\gamma} \left( (\epsilon_{\alpha}^* \epsilon_{\gamma} k^i_{\alpha} - \epsilon_{\beta}^* \epsilon_{\gamma} k^i_{\beta}) \Re \text{e} I_{\alpha\beta\gamma}(j, n) \right) \]

(8)

where \( \text{Den}(n, \omega, \Gamma) = \frac{m_e}{\hbar^2\hbar\omega} \frac{(E_n - E_g)^3}{h\omega - (E_n - E_g) - i\frac{\gamma}{2}} \). Finally, Eq. 9 can be rewritten in spherical coordinates, in a form that can be directly related to the multipoles of Table I:

\[ f_j^{DQ}(\omega) = \frac{i}{2} \sum_{l=1,2,3} \sum_{m \geq 0} \left( \frac{(-1)^m}{2} (T_{m}^{l+} T_{m}^{l-} - T_{m}^{l-} T_{m}^{l+}) \right) \]

(9)

Here the terms \( F^{l\pm} \), \( l = 1, 2, 3 \), correspond to the physical multipoles of Tables I and the terms \( T^{l\pm} \) are the corresponding polarization and wave-vector dependence. The knowledge of these latter terms is therefore useful experimentally in order to single out each single term of the multipolar development.

Consider any cartesian reference frame \( xyz \). In actual experiments, it is usually useful to choose such a frame in such a way that the \( z \) direction corresponds to an easy axis of the material, in order to simplify the \( F^{l\pm} \) tensor, or in such a way that some polarization and/or wave vectors components are zero, in order to simplify the \( T^{l\pm} \) term. In any of these reference frames, we have the following components for the geometrical x-ray parameters of the experiment:

a) The electric dipole is associated to the following components:

\[ n_z \sim \frac{3}{2\sqrt{15}} \left( (\epsilon_{\alpha}^* \epsilon_{\gamma} k^i_{\alpha} - \epsilon_{\beta}^* \epsilon_{\gamma} k^i_{\beta}) + \epsilon_{x}^* \epsilon_{y} k^i_{x} + \epsilon_{y}^* \epsilon_{z} k^i_{y} - \epsilon_{z}^* \epsilon_{x} k^i_{z} \right) + \frac{1}{\sqrt{15}} \left( 2 \epsilon_{x}^* \epsilon_{z} k^i_{x} + \epsilon_{y}^* \epsilon_{z} k^i_{y} - \epsilon_{z}^* \epsilon_{x} k^i_{z} \right) \]

(10)

\[ n_x \sim \frac{1}{2\sqrt{30}} \left( 4 \epsilon_{x}^* \epsilon_{y} k^i_{x} + 3 \epsilon_{y}^* \epsilon_{x} k^i_{y} + 3 \epsilon_{z}^* \epsilon_{x} k^i_{z} - 2 \epsilon_{x}^* \epsilon_{y} k^i_{y} + \epsilon_{x}^* \epsilon_{z} k^i_{y} - \epsilon_{y}^* \epsilon_{x} k^i_{z} \right) \]

(11)

\[ n_y \sim \frac{1}{2\sqrt{30}} \left( 4 \epsilon_{x}^* \epsilon_{y} k^i_{y} + 3 \epsilon_{y}^* \epsilon_{x} k^i_{x} + 3 \epsilon_{z}^* \epsilon_{x} k^i_{z} - 2 \epsilon_{x}^* \epsilon_{y} k^i_{z} + \epsilon_{x}^* \epsilon_{z} k^i_{z} - \epsilon_{y}^* \epsilon_{x} k^i_{z} \right) \]

(12)
b) The polar (magnetic) toroidal moment is associated to the following components:

\[ \Omega_z \sim \frac{3}{2\sqrt{15}} \left( (\epsilon_x^{\alpha\beta} \epsilon^{ij}_x + \epsilon_y^{\alpha\beta} \epsilon^{ij}_y)(k^i_x + k^o_x) + \epsilon_x^{\alpha\beta} \epsilon^{ij}_yk^i_x + \epsilon_y^{\alpha\beta} \epsilon^{ij}_yk^i_y + \epsilon_z^{\alpha\beta} \epsilon^{ij}_zk^i_z \right) + \frac{1}{\sqrt{15}} \left( 2\epsilon_x^{\alpha\beta} \epsilon^{ij}_x(k^i_x + k^o_x) - \epsilon_x^{\alpha\beta} \epsilon^{ij}_yk^i_x - \epsilon_y^{\alpha\beta} \epsilon^{ij}_yk^i_y - \epsilon_z^{\alpha\beta} \epsilon^{ij}_zk^i_z - \epsilon_z^{\alpha\beta} \epsilon^{ij}_yk^i_y \right) \]  

\[ (13) \]

\[ \Omega_x \sim \frac{1}{2\sqrt{30}} \left( (4\epsilon_x^{\alpha\beta} \epsilon^{ij}_x + 3\epsilon_y^{\alpha\beta} \epsilon^{ij}_y + 3\epsilon_z^{\alpha\beta} \epsilon^{ij}_z)(k^i_x + k^o_x) - 2(\epsilon_x^{\alpha\beta} \epsilon^{ij}_y k^i_x + \epsilon_y^{\alpha\beta} \epsilon^{ij}_yk^i_y + \epsilon_x^{\alpha\beta} \epsilon^{ij}_z k^i_z) + 3(\epsilon_y^{\alpha\beta} \epsilon^{ij}_x k^i_x + \epsilon_z^{\alpha\beta} \epsilon^{ij}_y k^i_y + \epsilon_y^{\alpha\beta} \epsilon^{ij}_z k^i_z) \right) \]  

\[ (14) \]

\[ \Omega_y \sim \frac{1}{2\sqrt{30}} \left( (4\epsilon_y^{\alpha\beta} \epsilon^{ij}_y + 3\epsilon_x^{\alpha\beta} \epsilon^{ij}_x + 3\epsilon_z^{\alpha\beta} \epsilon^{ij}_z)(k^i_y + k^o_y) - 2(\epsilon_x^{\alpha\beta} \epsilon^{ij}_y k^i_x + \epsilon_y^{\alpha\beta} \epsilon^{ij}_yk^i_y + \epsilon_x^{\alpha\beta} \epsilon^{ij}_z k^i_z) + 3(\epsilon_x^{\alpha\beta} \epsilon^{ij}_y k^i_x + \epsilon_z^{\alpha\beta} \epsilon^{ij}_y k^i_y + \epsilon_x^{\alpha\beta} \epsilon^{ij}_y k^i_z) \right) \]  

\[ (15) \]

c) The diagonal component \((l=2,m=0)\) of the axial toroidal quadrupole is associated to the following components of the x-ray tensor:

\[ L_z \Omega_z \sim \frac{1}{2} \left( (\epsilon_x^{\alpha\beta} \epsilon^{ij}_y + \epsilon_y^{\alpha\beta} \epsilon^{ij}_x)(k^i_x + k^o_x) + \epsilon_x^{\alpha\beta} \epsilon^{ij}_yk^i_y - \epsilon_y^{\alpha\beta} \epsilon^{ij}_yk^i_x - \epsilon_z^{\alpha\beta} \epsilon^{ij}_yk^i_x \right) \]  

\[ (16) \]

d) The diagonal component \((l=2,m=0)\) of the magnetic quadrupole is associated to the following components of the x-ray tensor:

\[ n_z \Omega_z \sim \frac{1}{2} \left( (\epsilon_x^{\alpha\beta} \epsilon^{ij}_y + \epsilon_y^{\alpha\beta} \epsilon^{ij}_x)(k^i_x + k^o_x) + \epsilon_x^{\alpha\beta} \epsilon^{ij}_yk^i_y - \epsilon_y^{\alpha\beta} \epsilon^{ij}_yk^i_x + \epsilon_x^{\alpha\beta} \epsilon^{ij}_yk^i_x \right) \]  

\[ (17) \]

e) The diagonal component \((l=3,m=0)\) of the electric octupole is associated to the following components of the x-ray tensor:

\[ n_z(3L_z^2 - L^2) \sim \frac{1}{\sqrt{10}} \left( (2\epsilon_x^{\alpha\beta} \epsilon^{ij}_x - \epsilon_y^{\alpha\beta} \epsilon^{ij}_y)(k^i_x - k^o_x) - (\epsilon_y^{\alpha\beta} \epsilon^{ij}_y + \epsilon_z^{\alpha\beta} \epsilon^{ij}_y)(k^i_y - k^o_y) \right) \]  

\[ (18) \]

f) The diagonal component \((l=3,m=0)\) of the polar toroidal octupole is associated to the following components of the x-ray tensor:

\[ \Omega_z(3L_z^2 - L^2) \sim \frac{1}{\sqrt{10}} \left( (2\epsilon_x^{\alpha\beta} \epsilon^{ij}_x - \epsilon_y^{\alpha\beta} \epsilon^{ij}_y)(k^i_x + k^o_x) - (\epsilon_y^{\alpha\beta} \epsilon^{ij}_y + \epsilon_z^{\alpha\beta} \epsilon^{ij}_y)(k^i_y + k^o_y) \right) \]  

\[ (19) \]

It should be noted that, in general, as shown in Ref. [5], magnetic and non-magnetic components for a given \((l,m)\) are transformed one into the other by the change \(k^o \rightarrow -k^o\).

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