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Frequency-dependent electric and magnetic multipole moments and Siegert's theorem

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Abstract. We discuss the definition of frequency-dependent electric and magnetic multipole moments. It is shown from classical electrodynamics that there are two types of magnetic multipole moments, $M_E(lm\omega)$ and $M_M(lm\omega)$, defined as moments of the magnetisation with an electric or magnetic spherical vector wave as weight function. The moment $M_E(lm\omega)$ contributes to the emission of electric multipole radiation. We define quantum-mechanical multipole operators in correspondence with the classical case and discuss Siegert's theorem.

1. Introduction

The main issue of this paper is the correct definition of frequency-dependent electric and magnetic multipole moments. Siegert's theorem (Siegert 1937, Sachs and Austern 1951) states that, to a good approximation, the transition amplitude for electric multipole radiation from a nucleus is determined by the matrix element of the electric multipole-moment operator

$$P(lm) = \int r^l Y^*_{lm}(\theta, \phi)\rho(\boldsymbol{r})\,\mathrm{d}\boldsymbol{r}, \tag{1.1}$$

where Y_{lm} is a spherical harmonic and $(\boldsymbol{r})$ is the charge density. The important feature which simplifies the calculation of transition amplitudes is that the operator $P(lm)$ has a form independent of the detailed dynamics of the nucleus. At higher photon energies the dynamics begins to play a role. Then the operator for electric multipole radiation can be written as a sum of two terms, customarily called the primary and secondary electric moments (Brennan and Sachs 1952), with the primary moment being given by an integral over the charge density with a frequency-dependent weight function, and the secondary moment depending on the detailed dynamics of the nucleus. If the second contribution can be regarded as small, then one has again a form of Siegert's theorem. In the literature there is, however, no agreement on how the above separation into two terms is to be made (Brennan and Sachs 1952, Waroquier *et al* 1975).

In this paper we demonstrate that the separation can be made unique by appealing to correspondence with the classical case. Already in classical electrodynamics electric multipole radiation can be shown to be radiated by a sum $\dot{P}_E(lm\omega) - \mathrm{i}\omega M_E(lm\omega)$, where $P_E(lm\omega)$ is a frequency-dependent electric multipole moment of the type (1.1) and $M_E(lm\omega)$ is a magnetic multipole moment. The latter is defined uniquely as a moment of the magnetisation with an electric vector wave as weight function. Magnetic multipole radiation is emitted by a magnetic moment $M_{\mathrm{M}}(lm\omega)$ defined as a moment of the magnetisation with a magnetic vector wave as weight function. Siegert's theorem holds in as far as the radiation emitted by $M_{\mathrm{E}}(lm\omega)$ can be neglected.

In §§ 2 and 3 we deal with the classical definition of multipole coefficients and of frequency-dependent electric and magnetic multipole moments. In § 4 we deal with the definition of multipole operators in quantum mechanics and discuss Siegert's theorem. Strictly speaking, the complete correspondence with the classical case can be established only for the Schrödinger Hamiltonian. For general Hamiltonian operators the question arises of which is the correct definition of the current density operator. This question will be studied in the following paper where we shall extend the correspondence with the classical case to general Hamiltonian operators.

2. Spherical vector wave expansion

We consider time-varying charge and current densities $\rho(\boldsymbol{r}, t)$ and $\boldsymbol{j}(\boldsymbol{r}, t)$ and an intrinsic magnetisation $\mathcal{M}(\boldsymbol{r}, t)$, which are localised around a fixed point in space. The variables are regarded as classical. We wish to describe the emitted electromagnetic radiation by means of a multipole expansion. We derive the electromagnetic fields from scalar and vector potentials,

$$\boldsymbol{E} = -\nabla - (1/c)\partial \boldsymbol{A}/\partial t, \qquad \boldsymbol{B} = \nabla \times \boldsymbol{A}, \tag{2.1}$$

and choose the Coulomb gauge $\nabla \cdot \boldsymbol{A} = 0$. Then Maxwell's equations in a vacuum are equivalent to

$$\nabla^2 \phi = -4\pi\rho, \qquad \nabla^2 \boldsymbol{A} - \frac{1}{c^2}\frac{\partial^2 \boldsymbol{A}}{\partial t^2} - \frac{1}{c}\frac{\partial \nabla}{\partial t} = -\frac{4\pi}{c}\mathcal{J}, \tag{2.2}$$

where $\mathcal{J}(\boldsymbol{r}, t)$ is the total current density,

$$\mathcal{J}(\boldsymbol{r}, t) = \boldsymbol{j}(\boldsymbol{r}, t) + c\nabla \times \mathcal{M}(\boldsymbol{r}, t). \tag{2.3}$$

Hence ϕ is given by the electrostatic Coulomb potential,

$$(\boldsymbol{r}, t) = \int \frac{(\boldsymbol{r}', t)}{|\boldsymbol{r} - \boldsymbol{r}'|} \, \mathrm{d}\boldsymbol{r}', \tag{2.4}$$

and using the continuity equation

$$\partial\rho/\partial t = -\nabla \cdot \boldsymbol{j} = -\nabla \cdot \mathcal{J}, \tag{2.5}$$

we find for the vector potential the inhomogeneous wave equation

$$\nabla^2 \boldsymbol{A} - \frac{1}{c^2}\frac{\partial^2 \boldsymbol{A}}{\partial t^2} = -\frac{4\pi}{c}\mathcal{J}_{\perp}, \tag{2.6}$$

where $\mathcal{J}_{\perp}$ is the transverse part of the total current density. We make a Fourier transformation with respect to time, and define

$$\boldsymbol{A}_{\omega}(\boldsymbol{r}) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \boldsymbol{A}(\boldsymbol{r}, t)\, \mathrm{e}^{\mathrm{i}\omega t} \, \mathrm{d}t, \tag{2.7}$$

and similarly for other quantities.

Far from the source the vector potential can be expanded in terms of outgoing spherical vector waves, so that asymptotically, for large r,

$$\boldsymbol{A}_\omega(\boldsymbol{r}) \approx \sum_{l=0}^{\infty} \sum_{m=-l}^{l} (a_{\mathrm{E}}(lm\omega)\boldsymbol{A}_{lm}^{\mathrm{E}+}(\boldsymbol{r}, \omega) + a_{\mathrm{M}}(lm\omega)\boldsymbol{A}_{lm}^{\mathrm{M}+}(\boldsymbol{r}, \omega)), \tag{2.8}$$

where $a_{\mathrm{E}}(lm\omega)$ and $a_{\mathrm{M}}(lm\omega)$ are the *electric and magnetic multipole coefficients*, and the spherical vector waves are given by

$$\boldsymbol{A}_{lm}^{\mathrm{E}+}(\boldsymbol{r}, \omega) = k^{-2}\nabla \times (h_l^{(1)}(kr)\boldsymbol{X}_{lm}(\theta, \phi)), \qquad \boldsymbol{A}_{lm}^{\mathrm{M}+}(\boldsymbol{r}, \omega) = (-\mathrm{i}/k)h_l^{(1)}(kr)\boldsymbol{X}_{lm}(\theta, \phi), \tag{2.9}$$

where $k = \omega/c$ is the wavenumber, $h_l^{(1)}(kr)$ is a spherical Hankel function and $\boldsymbol{X}_{lm}(\theta, \phi)$ is defined by

$$\boldsymbol{X}_{lm}(\theta, \phi) = [l(l+1)]^{-1/2}\boldsymbol{L}Y_{lm}(\theta, \phi), \tag{2.10}$$

where $\boldsymbol{L}$ is the differential operator $-\mathrm{i}\boldsymbol{r} \times \nabla$ and $Y_{lm}(\theta, \phi)$ are the spherical harmonics in the notation of Condon and Shortley (1935). We have normalised the solutions (2.9) such that the multipole coefficients are identical to those defined by Jackson (1962). They must be multiplied by $-\frac{1}{2}\mathrm{i}$ to obtain the corresponding coefficients of Blatt and Weisskopf (1952) (note that their source densities as defined by equations (3.1) and (3.2) on p 590 differ by a factor $\frac{1}{2}$ from ours). The solutions (2.9) are related by

$$\nabla \times \boldsymbol{A}_{lm}^{\mathrm{E}+} = \mathrm{i}k\boldsymbol{A}_{lm}^{\mathrm{M}+}, \qquad \nabla \times \boldsymbol{A}_{lm}^{\mathrm{M}+} = -\mathrm{i}k\boldsymbol{A}_{lm}^{\mathrm{E}+}, \tag{2.11}$$

and have the parity properties

$$\boldsymbol{A}_{lm}^{\mathrm{E}+}(-\boldsymbol{r}) = (-1)^{l+1}\boldsymbol{A}_{lm}^{\mathrm{E}+}(\boldsymbol{r}), \qquad \boldsymbol{A}_{lm}^{\mathrm{M}+}(-\boldsymbol{r}) = (-1)^{l}\boldsymbol{A}_{lm}^{\mathrm{M}+}(\boldsymbol{r}). \tag{2.12}$$

Corresponding solutions of the homogeneous wave equation are

$$\boldsymbol{A}_{lm}^{\mathrm{E}0}(\boldsymbol{r}, \omega) = k^{-2}\nabla \times (j_l(kr)\boldsymbol{X}_{lm}(\theta, \phi)), \qquad \boldsymbol{A}_{lm}^{\mathrm{M}0}(\boldsymbol{r}, \omega) = (-\mathrm{i}/k)j_l(kr)\boldsymbol{X}_{lm}(\theta, \phi), \tag{2.13}$$

where $j_l(kr)$ is the spherical Bessel function which is regular at the origin.

3. Electric and magnetic multipole moments

The inhomogeneous wave equation (2.6) can be solved by the method of Green functions, as indicated by Jackson (1962). Thus one finds for the multipole coefficients

$$\begin{aligned} a_{\mathrm{E}}(lm\omega) &= \frac{4\pi\mathrm{i}k^3}{c}\int \mathcal{J}_\omega(\boldsymbol{r})\cdot\boldsymbol{A}_{lm}^{\mathrm{E}0*}(\boldsymbol{r}, \omega)\,\mathrm{d}\boldsymbol{r}, \\ a_{\mathrm{M}}(lm\omega) &= \frac{4\pi\mathrm{i}k^3}{c}\int \mathcal{J}_\omega(\boldsymbol{r})\cdot\boldsymbol{A}_{lm}^{\mathrm{M}0*}(\boldsymbol{r}, \omega)\,\mathrm{d}\boldsymbol{r}, \end{aligned} \tag{3.1}$$

where we have performed an integration by parts in equations (16.88) and (16.89) of Jackson (1962). By Taylor expansion about the origin we decompose the current density into a sum of three terms,

$$\mathcal{J}_\omega(\boldsymbol{r}) = -\mathrm{i}\omega\boldsymbol{P}_\omega(\boldsymbol{r}) + c\nabla \times \boldsymbol{M}_\omega(\boldsymbol{r}) + c\nabla \times \mathcal{M}_\omega(\boldsymbol{r}), \tag{3.2}$$

where $\boldsymbol{P}_\omega(\boldsymbol{r})$ is the electric polarisation,

$$\boldsymbol{P}_\omega(\boldsymbol{r})=\int \boldsymbol{r}'\rho_\omega(\boldsymbol{r}')\sum_{n=0}^{\infty}\frac{(-1)^n}{(n+1)!}[(\boldsymbol{r}'.\nabla)^n\delta(\boldsymbol{r})]\,\mathrm{d}\boldsymbol{r}', \tag{3.3}$$

and $\boldsymbol{M}_\omega(\boldsymbol{r})$ is the orbital magnetisation,

$$\boldsymbol{M}_\omega(\boldsymbol{r})=\frac{1}{c}\int \boldsymbol{r}'\times\boldsymbol{j}_\omega(\boldsymbol{r}')\sum_{n=0}^{\infty}(-1)^n\frac{n+1}{(n+2)!}[(\boldsymbol{r}'.\nabla)^n\delta(\boldsymbol{r})]\,\mathrm{d}\boldsymbol{r}'. \tag{3.4}$$

Similarly the intrinsic magnetisation can be expanded as

$$\mathcal{M}_\omega(\boldsymbol{r})=\int \mathcal{M}_\omega(\boldsymbol{r}')\sum_{n=0}^{\infty}\frac{(-1)^n}{n!}[(\boldsymbol{r}'.\nabla)^n\delta(\boldsymbol{r})]\,\mathrm{d}\boldsymbol{r}'. \tag{3.5}$$

In order to prove the basic decomposition (3.2) we consider the integral

$$\int \boldsymbol{j}_\omega.\boldsymbol{F}\,\mathrm{d}\boldsymbol{r}=-\mathrm{i}\omega\int \boldsymbol{P}_\omega.\boldsymbol{F}\,\mathrm{d}\boldsymbol{r}+c\int(\nabla\times\boldsymbol{M}_\omega).\boldsymbol{F}\,\mathrm{d}\boldsymbol{r} \tag{3.6}$$

for an arbitrary test function $\boldsymbol{F}(\boldsymbol{r})$ and show that substitution of (3.3) and (3.4) leads to an identity. By partial integration and use of the continuity equation (2.5) one finds

$$\begin{aligned}-\mathrm{i}\omega\int \boldsymbol{P}_\omega.\boldsymbol{F}\,\mathrm{d}\boldsymbol{r}&=-\int x_\alpha(\partial_\beta j_{\omega\beta})\sum_{n=0}^{\infty}\frac{1}{(n+1)!}x_{\gamma_1}\ldots x_{\gamma_n}\,\partial_{\gamma_1}\ldots\partial_{\gamma_n}F_\alpha(0)\,\mathrm{d}\boldsymbol{r}\\&=\int j_{\omega\beta}\sum_{n=0}^{\infty}\frac{1}{(n+1)!}x_{\gamma_1}\ldots x_{\gamma_n}\,\partial_{\gamma_1}\ldots\partial_{\gamma_n}F_\beta(0)\,\mathrm{d}\boldsymbol{r}\\&\quad+\int x_\alpha j_{\omega\beta}\sum_{n=1}^{\infty}\frac{n}{(n+1)!}x_{\gamma_1}\ldots x_{\gamma_{n-1}}\,\partial_{\gamma_1}\ldots\partial_{\gamma_{n-1}}\,\partial_\beta F_\alpha(0)\,\mathrm{d}\boldsymbol{r}.\end{aligned} \tag{3.7}$$

Similarly

$$\begin{aligned}c\int(\nabla\times\boldsymbol{M}_\omega).\boldsymbol{F}\,\mathrm{d}\boldsymbol{r}&\\&=c\int \boldsymbol{M}_\omega.(\nabla\times\boldsymbol{F})\,\mathrm{d}\boldsymbol{r}\\&=\int x_\alpha j_{\omega\beta}\sum_{n=0}^{\infty}\frac{n+1}{(n+2)!}x_{\gamma_1}\ldots x_{\gamma_n}\,\partial_{\gamma_1}\ldots\partial_{\gamma_n}(\partial_\alpha F_\beta-\partial_\beta F_\alpha)(0)\,\mathrm{d}\boldsymbol{r}\\&=\int j_{\omega\beta}\sum_{n=0}^{\infty}\frac{n+1}{(n+2)!}x_{\gamma_1}\ldots x_{\gamma_{n+1}}\,\partial_{\gamma_1}\ldots\partial_{\gamma_{n+1}}F_\beta(0)\,\mathrm{d}\boldsymbol{r}\\&\quad-\int x_\alpha j_{\omega\beta}\sum_{n=0}^{\infty}\frac{n+1}{(n+2)!}x_{\gamma_1}\ldots x_{\gamma_n}\,\partial_{\gamma_1}\ldots\partial_{\gamma_n}\,\partial_\beta F_\alpha(0)\,\mathrm{d}\boldsymbol{r}.\end{aligned} \tag{3.8}$$

Adding (3.7) and (3.8) one sees that the last terms cancel while the first add up to $\int \boldsymbol{j}_\omega.\boldsymbol{F}\,\mathrm{d}\boldsymbol{r}$. The intrinsic magnetisation $\mathcal{M}_\omega(\boldsymbol{r})$ is included straightforwardly. The decomposition (3.2) is a continuum version of the atomic series expansion given by de Groot (1969).

The tensors multiplying $\nabla^n\delta(\boldsymbol{r})$ in (3.3–5) will be called the cartesian electric and magnetic multipole moments. From (3.3) one finds the identity

$$\int \boldsymbol{P}_\omega(\boldsymbol{r}).\boldsymbol{F}(\boldsymbol{r})\,\mathrm{d}\boldsymbol{r}=\int \boldsymbol{r}\rho_\omega(\boldsymbol{r}).\int_0^1\boldsymbol{F}(\lambda\boldsymbol{r})\,\mathrm{d}\lambda\,\mathrm{d}\boldsymbol{r} \tag{3.9}$$

for an arbitrary vector field $\boldsymbol{F}(\boldsymbol{r})$, and from (3.4)

$$\int (\nabla\times\boldsymbol{M}_\omega(\boldsymbol{r}))\boldsymbol{.}\boldsymbol{F}(\boldsymbol{r})\,\mathrm{d}\boldsymbol{r}=\frac{1}{c}\int(\boldsymbol{r}\times\boldsymbol{j}_\omega(\boldsymbol{r}))\boldsymbol{.}\int_0^1(\nabla\times\boldsymbol{F}(\lambda\boldsymbol{r}))\,\mathrm{d}\lambda\;\mathrm{d}\boldsymbol{r}. \tag{3.10}$$

Substituting (3.2) into (3.1), one can write the multipole coefficients as

$$\begin{aligned} a_{\mathrm{E}}(lm\omega)&=a_{\mathrm{E}}^{\mathrm{P}}(lm\omega)+a_{\mathrm{E}}^{\mathrm{M}}(lm\omega)+a_{\mathrm{E}}^{\mathcal{M}}(lm\omega),\\ a_{\mathrm{M}}(lm\omega)&=a_{\mathrm{M}}^{\mathrm{M}}(lm\omega)+a_{\mathrm{M}}^{\mathcal{M}}(lm\omega),\end{aligned} \tag{3.11}$$

with

$$\begin{aligned} a_{\mathrm{E}}^{\mathrm{P}}(lm\omega)&=4\pi\mathrm{i}k^3\int \boldsymbol{P}_\omega(\boldsymbol{r})\boldsymbol{.}\boldsymbol{E}_{lm}^{\mathrm{E0*}}(\boldsymbol{r},\omega)\,\mathrm{d}\boldsymbol{r},\\ a_{\mathrm{E}}^{\mathrm{M}}(lm\omega)&=4\pi\mathrm{i}k^3\int \boldsymbol{M}_\omega(\boldsymbol{r})\boldsymbol{.}\boldsymbol{B}_{lm}^{\mathrm{E0*}}(\boldsymbol{r},\omega)\,\mathrm{d}\boldsymbol{r},\\ a_{\mathrm{M}}^{\mathrm{M}}(lm\omega)&=4\pi\mathrm{i}k^3\int \boldsymbol{M}_\omega(\boldsymbol{r})\boldsymbol{.}\boldsymbol{B}_{lm}^{\mathrm{M0*}}(\boldsymbol{r},\omega)\,\mathrm{d}\boldsymbol{r},\end{aligned} \tag{3.12}$$

and similarly $a_{\mathrm{E,M}}^{\mathcal{M}}(lm\omega)$. In (3.11) we have used

$$a_{\mathrm{M}}^{\mathrm{P}}(lm\omega)=4\pi\mathrm{i}k^3\int \boldsymbol{P}_\omega(\boldsymbol{r})\boldsymbol{.}\boldsymbol{E}_{lm}^{\mathrm{M0*}}(\boldsymbol{r},\omega)\,\mathrm{d}\boldsymbol{r}=0, \tag{3.13}$$

noting that $\boldsymbol{E}_{lm}^{\mathrm{M0}}$ is tangential, whereas (3.9) shows that $\boldsymbol{P}$ is radial. The fact that $a_{\mathrm{E}}^{\mathrm{M}}(lm\omega)$ does not vanish shows that magnetic multipole moments can emit electric radiation.

Corresponding to the multipole coefficients (3.12) we define frequency-dependent spherical electric and magnetic E- and M-multipole moments by

$$\begin{aligned} P_{\mathrm{E}}(lm\omega,t)&=\int \boldsymbol{P}(\boldsymbol{r},t)\boldsymbol{.}\boldsymbol{E}_{lm}^{\mathrm{E0*}}(\boldsymbol{r},\omega)\,\mathrm{d}\boldsymbol{r},\\ M_{\mathrm{E}}(lm\omega,t)&=\int \boldsymbol{M}(\boldsymbol{r},t)\boldsymbol{.}\boldsymbol{B}_{lm}^{\mathrm{E0*}}(\boldsymbol{r},\omega)\,\mathrm{d}\boldsymbol{r},\\ M_{\mathrm{M}}(lm\omega,t)&=\int \boldsymbol{M}(\boldsymbol{r},t)\boldsymbol{.}\boldsymbol{B}_{lm}^{\mathrm{M0*}}(\boldsymbol{r},\omega)\,\mathrm{d}\boldsymbol{r},\end{aligned} \tag{3.14}$$

and similarly for the intrinsic magnetisation $\mathcal{M}(\boldsymbol{r},t)$. The multipole moments are dynamical variables defined for each value of ω. In order to investigate the multipole moments for small values of ω it is convenient to use (3.9) and (3.10) to transform the definitions (3.14). Thus one finds

$$P_{\mathrm{E}}(lm\omega,t)=\int \boldsymbol{r}\rho(\boldsymbol{r},t)\boldsymbol{.}\int_0^1 \boldsymbol{E}_{lm}^{\mathrm{E0*}}(\lambda\boldsymbol{r},\omega)\,\mathrm{d}\lambda\;\mathrm{d}\boldsymbol{r}, \tag{3.15}$$

where

$$\boldsymbol{E}_{lm}^{\mathrm{E0}}(\boldsymbol{r},\omega)=\frac{-1}{k[l(l+1)]^{1/2}}\{\nabla[(j_l(kr)+krj_l'(kr))Y_{lm}]+k^2\boldsymbol{r}j_l(kr)Y_{lm}]\}. \tag{3.16}$$

Taking the scalar product in (3.15) and performing the integration over λ, one can also write

$$P_{\mathrm{E}}(lm\omega,t)=\frac{-1}{k[l(l+1)]^{1/2}}\int \rho(\boldsymbol{r},t)(j_l(kr)+krj_l'(kr)+g_l(kr))Y_{lm}^*\,\mathrm{d}\boldsymbol{r}, \tag{3.17}$$

with

$$g_l(z)=\int_0^z xj_l(x)\,\mathrm{d}x. \tag{3.18}$$

Hence one finds, for small values of ω,

$$P_{\mathrm{E}}(lm\omega,t)\approx-\left(\frac{l+1}{l}\right)^{1/2}\frac{k^{l-1}}{(2l+1)!!}\int r^l Y^*_{lm}\rho(\boldsymbol{r},t)\,\mathrm{d}\boldsymbol{r}. \tag{3.19}$$

Similarly

$$M_{\mathrm{E}}(lm\omega,t)=\frac{-\mathrm{i}k}{c}\int(\boldsymbol{r}\times\boldsymbol{j}(\boldsymbol{r},t))\cdot\int_0^1\lambda\boldsymbol{A}^{\mathrm{M0}*}_{lm}(\lambda\boldsymbol{r})\,\mathrm{d}\lambda\,\mathrm{d}\boldsymbol{r}$$

$$=\frac{\mathrm{i}}{k^2c[l(l+1)]^{1/2}}\int\boldsymbol{j}(\boldsymbol{r},t)\cdot g_l(kr)\nabla Y^*_{lm}\,\mathrm{d}\boldsymbol{r}, \tag{3.20}$$

which becomes, for small values of ω,

$$M_{\mathrm{E}}(lm\omega,t)\approx\frac{\mathrm{i}}{c[l(l+1)]^{1/2}}\frac{k^l}{(l+3)(2l+1)!!}\int r^{l+2}\boldsymbol{j}(\boldsymbol{r},t)\cdot\nabla Y^*_{lm}\,\mathrm{d}\boldsymbol{r}. \tag{3.21}$$

Finally,

$$M_{\mathrm{M}}(lm\omega,t)=\frac{\mathrm{i}k}{c}\int(\boldsymbol{r}\times\boldsymbol{j}(\boldsymbol{r},t))\cdot\int_0^1\lambda\boldsymbol{A}^{\mathrm{E0}*}_{lm}(\lambda\boldsymbol{r})\,\mathrm{d}\lambda\,\mathrm{d}\boldsymbol{r}$$

$$=\frac{-1}{kc[l(l+1)]^{1/2}}\int\boldsymbol{j}(\boldsymbol{r},t)\cdot(\boldsymbol{r}\times\nabla j_l(kr)Y^*_{lm})\,\mathrm{d}\boldsymbol{r}. \tag{3.22}$$

For small values of ω,

$$M_{\mathrm{M}}(lm\omega,t)\approx\frac{-1}{c[l(l+1)]^{1/2}}\frac{k^{l-1}}{(2l+1)!!}\int r^lY^*_{lm}\nabla\cdot(\boldsymbol{r}\times\boldsymbol{j}(\boldsymbol{r},t))\,\mathrm{d}\boldsymbol{r}. \tag{3.23}$$

Note that M_{E} is smaller than M_{M}, approximately by a factor kR, where R is the dimension of the source. Expressions similar to (3.20–23) can be derived for $\mathcal{M}_{\mathrm{E}}(lm\omega,t)$ and $\mathcal{M}_{\mathrm{M}}(lm\omega,t)$.

By analogy with the moments (3.11) we also define

$$\mathcal{J}_{\mathrm{E}}(lm\omega,t)=\int\mathcal{J}(\boldsymbol{r},t)\cdot\boldsymbol{A}^{\mathrm{E0}*}_{lm}(\boldsymbol{r},\omega)\,\mathrm{d}\boldsymbol{r},\qquad \mathcal{J}_{\mathrm{M}}(lm\omega,t)=\int\mathcal{J}(\boldsymbol{r},t)\cdot\boldsymbol{A}^{\mathrm{M0}*}_{lm}(\boldsymbol{r},\omega)\,\mathrm{d}\boldsymbol{r}. \tag{3.24}$$

These are related to the moments defined above by

$$\begin{aligned}\mathcal{J}_{\mathrm{E}}(lm\omega,t)&=(\mathrm{i}/k)\,\mathrm{d}P_{\mathrm{E}}(lm\omega,t)/\mathrm{d}t+cM_{\mathrm{E}}(lm\omega,t)+c\mathcal{M}_{\mathrm{E}}(lm\omega,t),\\ \mathcal{J}_{\mathrm{M}}(lm\omega,t)&=cM_{\mathrm{M}}(lm\omega,t)+c\mathcal{M}_{\mathrm{M}}(lm\omega,t).\end{aligned} \tag{3.25}$$

In the next section we turn to quantum mechanics and compare the definition of electric and magnetic multipole operators with that of the classical dynamical variables.

4. Multipole operators in quantum mechanics

We consider a general Hamiltonian operator of the form

$$H = H^{\mathrm{At}}(\{\boldsymbol{p}_k - (e_k/c)\boldsymbol{A}(\boldsymbol{r}_k), \boldsymbol{r}_k, \boldsymbol{s}_k\}) + H^{\mathrm{Rad}} - \int \boldsymbol{\mathcal{M}}(\boldsymbol{r}) . \boldsymbol{B}(\boldsymbol{r})\, \mathrm{d}\boldsymbol{r}, \tag{4.1}$$

where H^{At} is the Hamiltonian for the atom or nucleus in interaction with the radiation field, H^{Rad} is the Hamiltonian for the free radiation field,

$$H^{\mathrm{Rad}} = \frac{1}{8\pi} \int (\boldsymbol{E}_{\perp}^2(\boldsymbol{r}) + \boldsymbol{B}^2(\boldsymbol{r}))\, \mathrm{d}\boldsymbol{r}, \tag{4.2}$$

and the intrinsic magnetisation $\boldsymbol{\mathcal{M}}(\boldsymbol{r})$ is defined by

$$\boldsymbol{\mathcal{M}}(\boldsymbol{r}) = \sum_j \mu_j \boldsymbol{s}_j \delta(\boldsymbol{r} - \boldsymbol{r}_j), \tag{4.3}$$

where the $\boldsymbol{s}_j$ are spin operators and the μ_j are values of the intrinsic magnetic moments. The vector potential $\boldsymbol{A}(\boldsymbol{r})$ has the expansion (Moskowski 1965)

$$\boldsymbol{A}(\boldsymbol{r}) = \sum_{\omega} \sum_{lm} \left(\frac{4\pi\hbar\omega^3}{Rc^2}\right)^{1/2} (\alpha_{\mathrm{E}}(lm\omega)\boldsymbol{A}_{lm}^{\mathrm{E}0}(\boldsymbol{r}, \omega) + \alpha_{\mathrm{M}}(lm\omega)\boldsymbol{A}_{lm}^{\mathrm{M}0}(\boldsymbol{r}, \omega) + \mathrm{HC}), \tag{4.4}$$

where R is the radius of a large sphere with reflecting boundary conditions and the first sum runs over the discrete frequencies $\omega_n = n(\pi/R)c$ with positive integers n. The operators $\alpha_{\mathrm{E,M}}(lm\omega)$, $\overset{\dagger}{\alpha}_{\mathrm{E,M}}(lm\omega)$ satisfy the commutation rules for creation and annihilation operators, e.g.

$$[\alpha_{\mathrm{E}}(lm\omega), \overset{\dagger}{\alpha}_{\mathrm{E}}(l'm'\omega')] = \delta_{ll'}\delta_{mm'}\delta_{\omega\omega'}. \tag{4.5}$$

It is assumed that the longitudinal Coulomb interaction is included in (4.1) as a potential energy term.

Since the interaction with the radiation field is weak, one can expand H^{At} in powers of $\boldsymbol{A}$. Thus we obtain

$$H = H_0 + V_1 + V_2 + \dots, \tag{4.6}$$

where

$$H_0 = H_0^{\mathrm{At}}(\{\boldsymbol{p}_k, \boldsymbol{r}_k, \boldsymbol{s}_k\}) + H^{\mathrm{Rad}}, \qquad V_1 = V_1^{\mathrm{orb}} - \int \boldsymbol{\mathcal{M}}(\boldsymbol{r}) . \boldsymbol{B}(\boldsymbol{r})\, \mathrm{d}\boldsymbol{r} \tag{4.7}$$

with

$$V_1^{\mathrm{orb}}(\boldsymbol{A}(\boldsymbol{r})) = -\sum_{j\alpha} \frac{e_j}{c} H_{1\alpha}^j(\{\boldsymbol{p}_k, \boldsymbol{r}_k, \boldsymbol{s}_k\}, A_\alpha(\boldsymbol{r}_j)). \tag{4.8}$$

By definition, the operator $H_{1\alpha}^j$ is linear in $A_\alpha(\boldsymbol{r}_j)$. To make the definition precise it is assumed that in writing out its form no use is made of the commutation relation between $\boldsymbol{r}_j$ and $\boldsymbol{p}_j$. In the following we shall restrict ourselves to first-order radiation processes determined by the term V_1. The transition probabilities for such processes are given by

$$W_{fi} = (2\pi/\hbar)|\langle f| V_1(\boldsymbol{A}(\boldsymbol{r}))|\mathrm{i}\rangle|^2 \rho(E), \tag{4.9}$$

where $|i\rangle$ and $|f\rangle$ are the initial and final states of $H_0^{\mathrm{At}} + H^{\mathrm{Rad}}$, and $\rho(E) = (R/\pi\hbar c)$ is the density of photon states per unit energy. Substituting from (4.4), we see that absorption processes involve the matrix element $\langle b| V_1^{\mathrm{orb}}(\boldsymbol{A}_{lm}^{\mathrm{E,M}0}(\boldsymbol{r}, \omega))|a\rangle$ where $|a\rangle$ and $|b\rangle$ are the initial and final eigenstate of H_0^{At}, whereas emission processes involve the matrix element $\langle b|V_1^{\mathrm{orb}}(\boldsymbol{A}_{lm}^{\mathrm{E,M0*}}(\boldsymbol{r},\omega))|a\rangle$.

We shall use the identity

$$\frac{\mathrm{i}}{\hbar}[H_0^{\mathrm{At}}, g(\boldsymbol{r}_j)] = \sum_{\alpha=1}^{3} H_1^j\left(\{\boldsymbol{p}_k, \boldsymbol{r}_k, \boldsymbol{s}_k\}, \frac{\partial g(\boldsymbol{r}_j)}{\partial r_{j\alpha}}\right), \tag{4.10}$$

where $g(\boldsymbol{r})$ is an arbitrary scalar function. To prove (4.10) one considers the transformation

$$\exp\left(-\frac{\mathrm{i}}{\hbar}\sum_j \lambda_j g(\boldsymbol{r}_j)\right)H_0^{\mathrm{At}}\exp\left(\frac{\mathrm{i}}{\hbar}\sum_j \lambda_j g(\boldsymbol{r}_j)\right) = H^{\mathrm{At}}\left(\left\{\boldsymbol{p}_k + \lambda_k\frac{\partial g(\boldsymbol{r}_k)}{\partial \boldsymbol{r}_k}, \boldsymbol{r}_k, \boldsymbol{s}_k\right\}\right)$$

$$= H_0^{\mathrm{At}} + \sum_{j\alpha}\lambda_j H_{1\alpha}^j\left(\{\boldsymbol{p}_k, \boldsymbol{r}_k, \boldsymbol{s}_k\}, \frac{\partial g(\boldsymbol{r}_j)}{\partial r_{j\alpha}}\right) + \mathrm{O}(\lambda^2). \tag{4.11}$$

Comparison with the expansion of the exponentials leads directly to (4.10).

Now suppose the electric spherical vector wave is written as a sum of two terms,

$$\boldsymbol{A}_{lm}^{\mathrm{E0}}(\boldsymbol{r},\omega) = \nabla f_{lm}(\boldsymbol{r},\omega) + \boldsymbol{g}_{lm}(\boldsymbol{r},\omega). \tag{4.12}$$

Then one can define corresponding operators $\dot{F}(lm\omega)$ and $G(lm\omega)$ by

$$\dot{F}(lm\omega) = V_1^{\mathrm{orb}}(\nabla f_{lm}(\boldsymbol{r},\omega)), \qquad G(lm\omega) = V_1^{\mathrm{orb}}(\boldsymbol{g}_{lm}(\boldsymbol{r},\omega)). \tag{4.13}$$

Applying the identity (4.10), one has

$$\dot{F}(lm\omega) = -\sum_j \frac{e_j}{c}\frac{\mathrm{i}}{\hbar}[H_0^{\mathrm{At}}, f_{lm}(\boldsymbol{r}_j,\omega)], \tag{4.14}$$

which suggests the definition of the operator

$$F(lm\omega) = -\sum_j \frac{e_j}{c} f_{lm}(\boldsymbol{r}_j,\omega) \tag{4.15}$$

and the interpretation of the dot as a Heisenberg time derivative defined by H_0^{At}. The matrix element for absorption now has a part which can be written

$$\langle b|V_1^{\mathrm{orb}}(\nabla f_{lm}(\boldsymbol{r},\omega))|a\rangle = (\mathrm{i}/\hbar)(E_b - E_a)\langle b|F(lm\omega)|a\rangle, \tag{4.16}$$

where $E_{a,b}$ are energy levels of the unperturbed atom. The calculation of the matrix element of $F(lm\omega)$ is obviously much simpler than that of $V_1^{\mathrm{orb}}(\boldsymbol{A}_{lm}^{\mathrm{E0}}(\boldsymbol{r},\omega))$, since $F(lm\omega)$ is a sum of single-particle operators and has a form which is independent of the dynamics. If the matrix element of $G(lm\omega)$ can be argued to be small, then the calculation of transition probabilities for electric multipole radiation is much simplified.

In the low-frequency limit, $\boldsymbol{A}_{lm}^{\mathrm{E0}}(\boldsymbol{r},\omega)$ is in fact a pure gradient, as is evident from the form

$$\boldsymbol{A}_{lm}^{\mathrm{E0}}(\boldsymbol{r},\omega) = \frac{\mathrm{i}}{k^2[l(l+1)]^{1/2}}\left\{\nabla[(j_l(kr) + krj_l'(kr))Y_{lm}] + k^2\boldsymbol{r}j_l(kr)Y_{lm}\right\}. \tag{4.17}$$

In the limit $\omega \to 0$ the last term can be neglected and $\boldsymbol{A}_{lm}^{\mathrm{E0}}(\boldsymbol{r},\omega)$ can be approximated by

$$\boldsymbol{A}_{lm}^{\mathrm{E0}}(\boldsymbol{r},\omega) \approx \left(\frac{l+1}{l}\right)^{1/2}\frac{\mathrm{i}k^{l-2}}{(2l+1)!!}\nabla(r^l Y_{lm}) \qquad (\omega \to 0). \tag{4.18}$$

Hence the above state of affairs prevails, and the calculation of matrix elements for electric multipole radiation reduces to calculating those of

$$F(lm) = -\left(\frac{l+1}{l}\right)^{1/2} \frac{\mathrm{i}k^{l-2}}{(2l+1)!!} \sum_j \frac{e_j}{c} (r^l Y_{lm})_{r=r_j}. \tag{4.19}$$

This is known as Siegert's theorem (Siegert 1937, Sachs and Austern 1951).

At higher frequencies the separation into two terms in (4.12) is not unique. Brennan and Sachs (1952) have used the form (4.17) and define

$$F(lm\omega) = -\frac{\mathrm{i}}{k^2[l(l+1)]^{1/2}} \sum_j \frac{e_j}{c} [(j_l(kr) + krj_l'(kr)) Y_{lm}]_{r=r_j} \tag{4.20}$$

as the primary electric multipole moment operator. They regard

$$G(lm\omega) = V_1^{\mathrm{orb}}\left(\frac{\mathrm{i}}{[l(l+1)]^{1/2}} r j_l(kr) Y_{lm}\right) \tag{4.21}$$

as the time derivative of a secondary electric multipole moment operator. On the other hand, Waroquier *et al* (1975) make a different splitting and use

$$f_{lm}(\boldsymbol{r}, \omega) = (\mathrm{i}/k^2)[(l+1)/l]^{1/2} j_l(kr) Y_{lm} \tag{4.22}$$

for the primary operator and the remainder for the secondary one. If one neglects $G(lm\omega)$ in either case, one clearly finds two different approximate values for the transition probability. In the low-frequency limit the two values agree.

Here we propose to make the separation in (4.12) unique by appealing to correspondence with the classical case. This suggests that the splitting is made in such a way that the first part corresponds to the electric multipole operator $P_{\mathrm{E}}(lm\omega)$ and the second part to the magnetic E-multipole operator $M_{\mathrm{E}}(lm\omega)$. Instead of (4.17) we write

$$\boldsymbol{A}_{lm}^{\mathrm{E0}}(\boldsymbol{r}, \omega) = \frac{\mathrm{i}}{k^2[l(l+1)]^{1/2}} \{\nabla[(j_l(kr) + krj_l'(kr) + g_l(kr)) Y_{lm}] - g_l(kr) \nabla Y_{lm}\} \tag{4.23}$$

where $g_l(kr)$ has been defined in (3.18). Now the operator $F(lm\omega)$ becomes

$$F(lm\omega) = -\frac{\mathrm{i}}{k^2 c[l(l+1)]^{1/2}} \sum_j e_j [(j_l(kr) + krj_l'(kr) + g_l(kr)) Y_{lm}]_{r=r_j}. \tag{4.24}$$

Slightly modifying the prefactor and taking the Hermitian conjugate, we define the electric multipole operator as

$$P_{\mathrm{E}}(lm\omega) = \mathrm{i}\omega F_{\mathrm{E}}^{\dagger}(lm\omega)$$

or

$$P_{\mathrm{E}}(lm\omega) = -\frac{1}{k[l(l+1)]^{1/2}} \sum_j e_j [(j_l(kr) + krj_l'(kr) + g_l(kr)) Y_{lm}^*]_{r=r_j}. \tag{4.25}$$

Comparison with (3.17) shows that this definition corresponds exactly with the classical expression. The operator $G(lm\omega)$ which follows from (4.23) is given by

$$G(lm\omega) = V_1^{\mathrm{orb}}\left(\frac{-\mathrm{i}}{k^2[l(l+1)]^{1/2}} g_l(kr) \nabla Y_{lm}\right). \tag{4.26}$$

Comparison with (3.20) for the case where H_0^{At} is given by the Schrödinger Hamiltonian suggests that one can write

$$G(lm\omega) = -M_{\mathrm{E}}^{\dagger}(lm\omega), \tag{4.27}$$

where the magnetic E-moment operator is defined by

$$M_{\mathrm{E}}(lm\omega)=\frac{\mathrm{i}}{k^2c[l(l+1)]^{1/2}}\int \boldsymbol{j}_0(\boldsymbol{r})\cdot g_l(kr)\nabla Y^*_{lm}\,\mathrm{d}\boldsymbol{r}, \tag{4.28}$$

where $\boldsymbol{j}_0(\boldsymbol{r})$ is the current density operator corresponding to the unperturbed Hamiltonian H_0^{At}. Again this agrees exactly with the classical definition (3.20). That (4.28) is in fact the correct definition for general Hamiltonian operators H_0^{At} will be shown in the following paper (Meister and Felderhof 1980). Using (4.13), (4.16), (4.24), (4.25) and (4.27) we find that the matrix element for emission of electric multipole radiation is given by

$$\langle b|V_1(\boldsymbol{A}^{\mathrm{E0}*}_{lm}(\boldsymbol{r},\omega))|a\rangle = \frac{E_t-E_a}{\hbar\omega}\langle b|P_{\mathrm{E}}(lm\omega)|a\rangle-\langle b|M_{\mathrm{E}}(lm\omega)|a\rangle-\langle b|\mathcal{M}_{\mathrm{E}}(lm\omega)|a\rangle. \tag{4.29}$$

Since, for emission $E_b-E_a=-\hbar\omega$, we may write, for matrix elements between states satisfying this condition,

$$\langle b|V_1(\boldsymbol{A}^{\mathrm{E0}*}_{lm}(\boldsymbol{r},\omega))|a\rangle = -\langle b|P_{\mathrm{E}}(lm\omega)+M_{\mathrm{E}}(lm\omega)+\mathcal{M}_{\mathrm{E}}(lm\omega)|a\rangle \qquad (E_b-E_a=-\hbar\omega). \tag{4.30}$$

Neglect of the term with $G(lm\omega)$ now has the clear physical significance that the contribution from the magnetic E-multipole moment $M_{\mathrm{E}}(lm\omega)$ is omitted. If this is done, and if the contribution from $\mathcal{M}_{\mathrm{E}}(lm\omega)$ is also omitted, one is left with the matrix element of $P_{\mathrm{E}}(lm\omega)$ given by (4.25), which is a simple operator independent of the dynamics. Siegert's theorem can be generalised to the frequency-dependent multipole moment operators and now reads: the transition amplitudes for emission and absorption of electric multipole radiation can, to a good approximation be calculated from the matrix elements of the frequency-dependent electric multipole moment operator $P_{\mathrm{E}}(lm\omega)$. How good the approximation is must be estimated from a more detailed calculation. For dipole transitions in nuclei with photon energies of the order of 10 MeV, such a calculation shows that the contribution from the magnetic E-moment M_{E} to the transition probability is of the order of one per cent or less. However, for the nuclear photon effect, where higher photon energies come into play, the contribution from $M_{\mathrm{E}}(lm\omega)$ can be appreciable. For a He^4 nucleus and a photon energy of 200 MeV the contribution is about ten per cent. The contribution from the intrinsic moment $\mathcal{M}_{\mathrm{E}}(lm\omega)$ is even larger.

Finally we consider the magnetic M-multipole moment operator $M_{\mathrm{M}}(lm\omega)$. Comparison with (4.23), (4.26) and (4.27) suggests the definition

$$M_{\mathrm{M}}(lm\omega)=-V_1^{\mathrm{orb}}(\boldsymbol{A}^{\mathrm{M0}*}_{lm}(\boldsymbol{r},\omega)). \tag{4.31}$$

For the Schrödinger Hamiltonian this can be written

$$M_{\mathrm{M}}(lm\omega)=(1/c)\int \boldsymbol{j}_0(\boldsymbol{r})\cdot\boldsymbol{A}^{\mathrm{M0}*}_{lm}(\boldsymbol{r})\,\mathrm{d}\boldsymbol{r}, \tag{4.32}$$

in exact agreement with the classical definition (3.14). In the following paper we show that (4.32) is also valid for more general Hamiltonian operators. The matrix element for emission of magnetic multipole radiation can be written

$$\langle b|V_1(\boldsymbol{A}_{lm}^{\mathrm{M}0*}(\boldsymbol{r},\omega))|a\rangle = -\langle b|M_{\mathrm{M}}(lm\omega)+\mathscr{M}_{\mathrm{M}}(lm\omega)|a\rangle \tag{4.33}$$

in analogy to (4.30).

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