Spherical tensor approach to multipole expansions.

II. Magnetostatic interactions\(^1,2\)

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The magnetic field due to a given current distribution, the interaction energy of a current distribution with an arbitrary external magnetic field, and the magnetostatic interaction energy between two current distributions are decomposed into multipolar components using spherical harmonic expansions. Diamagnetic interactions and the spin contributions to the multipole expansions are also discussed.

1. Introduction

Magnetic multipole moments higher than the dipole are usually discussed in connection with the theory of multipole radiation which involves the theory of vector spherical harmonics (Blatt and Weisskopf 1952; Schwartz 1955; Jackson 1962; Brink and Satchler 1962; Bohr and Mottelson 1969). A simple and general treatment of magnetostatic problems does not appear to exist although some special topics have been discussed (Jackson 1962). Examples of phenomena requiring higher magnetic multipole moments for their interpretation include the hyperfine interaction between the nuclear magnetic octupole moment and the electronic field gradient (Jaccarino et al. 1954; Kusch and Eck 1954; Daly and Holloway 1954), nuclear electromagnetic transition probabilities (Bohr and Mottelson 1969), and chemical shifts induced by polyatomic molecules in nuclear magnetic resonance spectroscopy (Stiles 1975) and references therein.

In this paper the magnetic field outside a current distribution (Sect. 2), the interaction energy of a current distribution with an arbitrary external field (Sect. 3), and the interaction energy between two current distributions (Sect. 4) are decomposed into multipolar components. The analogous electrostatic problems are discussed in the preceding paper (Gray 1976).

We also discuss the diamagnetic energy of interaction between a current distribution and an external field (Sect. 3) and modifications to the previous results which occur if the charged particles have intrinsic spin and associated magnetic dipole moments (Sect. 5). Most of the derivations and some of the results appear to be new.

2. The Magnetic Field Outside a Stationary Current Distribution

We consider a stationary \((\nabla \cdot \mathbf{J} = 0)\) current density distribution \(\mathbf{J}(\mathbf{R})\) which, for simplicity, we take to be discrete:

\[
\mathbf{J}(\mathbf{R}) = \sum_i e_i \mathbf{v}_i \delta(\mathbf{R} - \mathbf{r}_i)
\]

where \(\mathbf{v}_i\) is the velocity and \(e_i\) the charge of particle \(i\) at the point \(\mathbf{r}_i\) relative to an arbitrary origin in the distribution. In the Coulomb gauge \((\nabla \cdot \mathbf{A} = 0)\) the vector potential \(\mathbf{A}(\mathbf{R})\) arising from the current \(\mathbf{J}\) satisfies \(\nabla^2 \mathbf{A} = -4\pi \mathbf{J}/c\) in Gaussian units.

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The contribution to $A$ from particle $i$ is therefore

$$A_i(R) = (e_i v_i/c)$$

where $r = |R - r_i|$ is the distance between the field point and particle $i$. Although $A_i$ does not satisfy $\nabla \cdot A_i = 0$ we show in the Appendix that the total potential $A = \sum_i A_i$ satisfies $\nabla \cdot A = 0$.

An arbitrary vector field $A(R)$ can be derived from three scalar fields $\psi(R)$, $\chi(R)$, and $\phi(R)$ as follows (Brink and Satchler 1962):

$$A = L \psi + (\nabla \times L) \chi + \nabla \phi$$

where the operator $L = -i (R \times \nabla)$ is familiar from angular momentum theory. Since $\nabla^2$ commutes with $L$ and $\nabla$ we can ensure that $A(R)$ is a harmonic function in source-free regions of space by choosing $\psi(R)$, $\chi(R)$, and $\phi(R)$ to be harmonic in these regions. The gauge condition $\nabla \cdot A = 0$ fixes $\phi = 0$. Because the second term in [3] makes no contribution to the magnetic field $B = \nabla \times A$ at source-free field points, we can choose $\chi(R) = 0$. Outside the sources we can therefore write

$$A(R) = L \psi(R) \quad \text{(source-free regions only)}$$

Note that [4] implies that $A$ is perpendicular to $R$ and is divergence-free. From the operator identity $L \times L = iL$ we find from [4] that

$$A = -i L \times A = -L \times (L \times A) = L^2 A - L (L \cdot A) + A$$

and hence

$$L^2 A = L (L \cdot A)$$

Expanding the potential [2] in spherical harmonics using the one-centre result for $r^{-1}$ (see, e.g., Appendix VI of Brink and Satchler (1962) or Gray (1976)) we find

$$A(R) = (e_i v_i/c) \sum_{lm} r_i L_{lm} C_{lm}^{(\omega_i)} \phi_l(\Omega) / r^{l+1}$$

The $l = 0$ term can be omitted because of the absence of magnetic monopoles. We now rewrite [6], using [5] and $L^2 C_{lm} = (l + 1) C_{lm}$, as

$$A(R) = (e_i v_i/c) \sum_{lm} \frac{L_i v_i \cdot L}{(l + 1)} r_i L_{lm} C_{lm}^{(\omega_i)} / r^{l+1}$$

Because

$$\sum_{m} C_{lm}^{(\omega)} C_{lm}^{(\Omega)} = \frac{(l + 1)}{(2l + 1)}$$

is invariant under rotations we have

$$\sum_{m} C_{lm}^{(\omega)} C_{lm}^{(\Omega)} = 0$$

where $L_i = -i (r_i \times \nabla)$. Using [8] and the fact that $L_i$ commutes with $L$ we find

$$A(R) = -(e_i v_i/c) \sum_{lm} \frac{v_i \cdot L_i}{(l + 1)} r_i L_{lm} C_{lm}^{(\omega_i)} / r^{l+1} \times L C_{lm}^{(\Omega)} / r^{l+1}$$

The total vector potential $A$ due to all the charges in the distribution is thus the sum of multipole contributions

$$A(R) = \sum_{l>0} A_l(R)$$

where

$$A_l(R) = -(e_i v_i/c) \sum_{lm} \frac{v_i \cdot L}{(l + 1)} r_i L_{lm} C_{lm}^{(\omega_i)} / r^{l+1} \times L C_{lm}^{(\Omega)} / r^{l+1}$$

and the spherical components $M_{lm}$ of the magnetic multipole moment of order $l$ are defined by

$$M_{lm} = -(l + 1)^{-1} \sum_{i} (e_i v_i/c) (v_i \cdot (L_i r_i) \times L_i) r_i L_{lm} C_{lm}^{(\omega_i)}$$

By rearranging the triple product $v_i \cdot (r_i \times \nabla)$ and introducing the momentum $p_i = m_i v_i$ we see that the definition [12] can also be written as

$$M_{lm} = (l + 1)^{-1} \sum_{i} (e_i m_i/c) (v_i \cdot (p_i \times L_i) \times (r_i \times L_i)) r_i L_{lm} C_{lm}^{(\omega_i)}$$

For a continuous current distribution [13] is replaced by

$$M_{lm} = \frac{1}{c} (l + 1)^{-1} \int dr \ (r \times J(r)) \cdot \nabla r^l C_{lm}^{(\omega)}$$

Using Gauss's theorem and $\nabla \cdot J = 0$ we obtain yet another form from [14]

$$M_{lm} = \frac{1}{c} (l + 1)^{-1} \int dr \ r^l C_{lm}^{(\omega)} r \cdot (\nabla \times J(r))$$

A discussion of the properties of the $M_{lm}$ parallels that given for the electrostatic multipole moments
In Gray (1976). In particular we note that there are, in general, \((2l + 1)\) independent components of order \(l\), the parity is \((-1)^{l+1}\), and

\[ M_{lm} = (-1)^l m_{lm}, \]

where \(m \equiv -m\). Note that using \(\mathbf{r} \cdot \mathbf{C}_{lm} = r_m\) and \(V \mathbf{r} = \mathbf{I}\) in (13) gives the standard Cartesian form

\[ M_1 = \sum_i \left( \frac{e_i}{2e} \right) (r_i \times v_i) = m \]

for the dipole moment.

For dipole moments with axial symmetry, such as the nitric oxide molecule, (13) can be simplified to

\[ M_{lm} = M_l C_{lm}(\omega) \]

where \(\omega\) is the orientation of the symmetry axis and

\[ M_l = (l + 1)^{-1} \]

\[ \times \sum_i \left( \frac{e_i m e}{c^2} \right) (r_i \times p_i) \cdot \mathbf{v}_r r_i^l P_l,(\cos \theta) \]

is the scalar magnetic multipole of order \(l\), and is calculated in the body-fixed frame with the polar axis along the symmetry axis. The relation (18) can be proved in the same way as the corresponding electrostatic result (Gray 1976). The \(M_l\) are zero for even values of \(l\) if the current distribution has a centre of inversion.

The magnetic field \(\mathbf{B}(R) = \nabla \times \mathbf{A}(R)\) follows from (10) and (11) as the sum of multipole contributions

\[ B_l(R) = -\frac{l}{l+1} \sum_m M_{lm}(\nabla \times L) C_{lm}(\Omega)^*/R^{l+1} \]

From the operator identity

\[ \nabla \times L = -i R \nabla^2 + i \mathbf{V}(R \cdot \mathbf{V} + 1) \]

and \(\nabla^2 C_{lm}(\Omega)/R^{l+1} = 0\), \((R \neq 0)\), we have

\[ (\nabla \times L) C_{lm}(\Omega)/R^{l+1} = -i \nabla C_{lm}(\Omega)/R^{l+1}, \quad (R \neq 0) \]

which, when substituted in (20) gives

\[ B_l(R) = -\sum_m M_{lm} \nabla C_{lm}(\Omega)^*/R^{l+1} \]

We note that (22) indicates the existence of another magnetic scalar potential in source-free regions, defined by \(\Phi = -\nabla \phi\), where

\[ \Phi(R) = \sum_m M_{lm} C_{lm}(\Omega)^*/R^{l+1} \]

The scalar potential \(\Phi(R)\) is the one usually introduced in magnetostatic problems.

3. Interaction of a Current Distribution with an External Magnetic Field

The interaction energy between a system of particles (with charges \(e_i\), canonical momenta \(p_i\), and positions \(r_i\)) and an applied magnetic vector potential \(\mathbf{A}(r)\) is (Van Vleck 1932)

\[ V = -\sum R e_i p_i \cdot \mathbf{A}(r_i) + \sum \frac{e_i^2}{2 m_i c^2} A(r_i)^2 \]

\[ \equiv V(1) + V(2) \]

As in the previous section we derive \(\mathbf{A}\) from a scalar potential \(\psi\), using \(\mathbf{A}(r) = \nabla \psi(r)\), where \(\psi\) is harmonic in source-free regions. We expand \(\psi(r) = \sum_{lm} \psi_{lm} r^l C_{lm}(\omega)^*\)

and obtain

\[ A_l(R) = \sum_{lm} \psi_{lm} L r^l C_{lm}(\omega)^* \]

where, once again, the \(l = 0\) term makes no contribution.

The expansion coefficients \(\psi_{lm}\) can be related to the magnetic field and its gradients at the (arbitrary) origin in the distribution. Using the identity (20) and the fact that \(\nabla^2 C_{lm}(\omega) = 0\) we find

\[ (\nabla \times L) r^l C_{lm}(\omega) = i(l + 1) \nabla r^l C_{lm}(\omega) \]

Calculating \(\mathbf{B}(r)\) from (26) using \(\mathbf{B} = \nabla \times \mathbf{A}\) and (27) we get

\[ B_l(R) = \sum_{lm} B_{lm} \nabla r^l C_{lm}(\omega)^* \]

where the expansion coefficients for \(\mathbf{B}\) are given by

\[ B_{lm} = i(l + 1) \psi_{lm} \]

A comparison of (28) with the Taylor series expansion

\[ \mathbf{B}(r) = \mathbf{B}_0 + r \cdot (\nabla \mathbf{B})_0 + \frac{1}{2} r \cdot r : (\nabla \nabla \mathbf{B})_0 + \ldots \]

gives

\[ B_{1m} = (B_{m0}), B_{2m} = 6^{-1/2} \sum_{lm} C(112; \mu \nu m) \nabla_\mu B_\nu_0 \]

and so on, where \(C(1, 2; l; m_1, m_2, m)\) denotes a Clebsch–Gordan coefficient and \(\nabla_m\) and \(B_m\) are the spherical components of the gradient and field vectors \(\mathbf{V}\) and \(\mathbf{B}\): \(\nabla_0 = \nabla_x\) and \(\nabla_{\pm 1} = \pm 2^{-1/2}(\nabla_x \pm i \nabla_y)\).
Substituting $\psi_{l,m}$ from [29] into [26] gives
\[ V(1) = \frac{i}{4 \xi} \sum_{l,m} \frac{e_l}{m_c} B_{l,m} \sum_{l + 1} P_{l+1} \cdot L_{l+1} C_{l,m}(\Omega) \]
Thus, the first order interaction can be written as a sum of multipolar contributions $\sum_{l>0} V_l$, where
\[ V_l = -\sum_{m} M_{l,m} B_{l,m} \]
and the multipole moments $M_{l,m}$ are those defined by [13].
From [31] we see that, as in the electric case, the dipole interacts with the field, the quadrupole with the field gradient, etc.
The diamagnetic term in [24] can also be expanded in spherical harmonics using
\[ A^2 = L^2 \psi \cdot L^2 \psi = L^2 \psi^2 - \psi L^2 \psi \]
We find
\[ V(2) = -\frac{e^2}{4 \xi m_c^2} \sum_{l \geq 2} r_{l+1}^{l+1} C(l' l 000) \]
\[ \times \left[ \frac{L(L + 1)}{(l + 1)(l + 1)} - \frac{2l'}{l + 1} \right] \]
\[ \times \sum_{l} B_{l,m}(l') C_{l,m}(\Omega) \]
where
\[ B_{l,m}(l') = \sum_{m'} C(l'l; mm'M) B_{l,m} B_{l',m'} \]
As a simple example of an application of [33] we find that the magnetic dipole susceptibility defined by $\langle V(2) \rangle = -(1/2) \chi B_0^2$ of an s-state diamagnetic atom is given by the Langevin expression
\[ \chi = -(e^2/6m_c^2) \sum_{l} \langle r_{l+1}^2 \rangle \]

4. Multipolar Interactions Between Two Current Distributions

Consider two nonoverlapping stationary current distributions 1 and 2 centred about origins $O_1$ and $O_2$ which are connected by the vector $R = (\Omega \Omega)$ from $O_1$ to $O_2$. The distance between a typical pair of charges $e_1$ and $e_2$ with velocities $v_1$ and $v_2$ in the distributions 1 and 2 is denoted by $r_{12} = |R + r_2 - r_1|$, where $r_i \equiv (r_i, \omega_i)$ is the position of $e_i$ relative to $O_i$ and $r_2$ the position of $e_2$ relative to $O_2$. The orientations $\omega_1$, $\omega_2$, and $\Omega$ refer to an arbitrary polar axis.

The leading order contribution from $e_1$ and $e_2$ to the magnetostatic interaction follows from [24] as
\[ V_{12} = -(e_2/e_c) v_2 \cdot A_{12}(r_{12}) \]
where $A_{12}(r_{12})$ is the potential due to $e_1$ at the position $r_{12} = R + r_2$ of $e_2$ relative to $O_1$. Using [2] this becomes
\[ V_{12} = -\frac{e_1 e_2 v_1 \cdot v_2}{c^2 r_{12}} \]
Strictly speaking the Darwin correction (Darwin 1920)
\[ \frac{1}{2} \left( \frac{e_1 e_2}{c^2} \right) \left[ \frac{(v_1 \cdot r_{12})(v_2 \cdot r_{12})}{r_{12}^2} - \frac{v_1 \cdot v_2}{r_{12}} \right] \]
which is of the same order in $v/c$ and in $r_{12}^{-1}$, should be added to [36]. However, this term vanishes (Oppenheimer 1970) when summed over all the charges in the stationary distributions. A simple proof of this point is given in the Appendix.
We now use the result
\[ L^2_1 L^2_2 V_{12} = -(e_1 e_2/c^2)(v_1 \cdot L_1)(v_2 \cdot L_2)(L_1 \cdot L_2)^{-1} \]
which can be obtained using condition [5] for $A_{12}(r_{12})$, namely
\[ L^2_2 e_1 v_1 / r_{12} = L^2_2 e_1 v_1 / r_{12} \]
where $L^2_2 = -i r_{12} \times V_{12}$. The derivation of [37] from [38] uses the rotational invariance of $r_{12}^{-1}$ in the form $(L_1 \cdot L_2)r_{12}^{-1} = 0$. (Again, [37] is strictly only valid when we sum both sides over all the charges of the two distributions.)
Substituting the two-centre expansion (Gray 1968, 1976) for $r_{12}^{-1}$ in both sides of [37] we find
\[ V_{12} = -\frac{e_1 e_2}{c^2} \sum_{l \geq 2} A_{1+2; 1+2} (v_1 \cdot L_1) \]
\[ \times \left( \frac{v_2 \cdot L_2}{l(l+1)} \right) (L_1 \cdot L_2) \chi(l_1 l_2) \]
where $l = l_1 + l_2$.
\[ A_{l+2; l+2} = (\xi)^{1/2} \left( \frac{(2l)!}{(2l_1)! (2l_2)!} \right)^{1/2} \]
\[ \chi(l_1 l_2) = \sum_{m_1 m_2} C(l_1 l_2; m_1 m_2) \]
\[ \times C_{l_1 m_1}(\omega_1) C_{l_2 m_2}(\omega_2) C_{l m}(\Omega) \]
and the dash on the summation sign indicates the exclusion of \( I_1 = I_2 = 0 \) terms from the sum. Since \( \chi(l_1l_2l) \) is rotationally invariant we have \((L_1 + L_2 + L)^2 \chi = (L_1 + L_2 + L)^2 \chi = 0\). Using these relations and the three results of the type \( L^2 \chi = l_1(l_1 + 1) \chi \) we find

\[ (L_1 \cdot L_2) \chi(l_1l_2l) = l_1l_2 \chi(l_1l_2l) \]

From [39] and [42] we see that the total magnetostatic interaction energy between the two distributions can be written as a sum of multipolar contributions

\[ V = \sum_{l_1l_2} V_{l_1l_2} \]

where

\[ V_{l_1l_2} = \left( A_{l_1l_2} / R^{l_1l_2} \right) \sum_{m_1m_2m} C(l_1l_2l; m_1m_2m) \]

\[ \times M_{1m_1} M_{2m_2} C_{lm}(\Omega) \]

and the multipole moments \( M_{lm} \) are defined by [12].

Equation 43 gives the general magnetostatic interaction energy between the multipole moments of orders \( l_1 \) and \( l_2 \) of the distributions 1 and 2, \( V_{l_1l_2} \) is the dipole–dipole term, \( V_{12} \) the dipole–quadrupole term, and so on. Note that, as for the electrostatic case, only terms corresponding to \( I = l_1 + l_2 \) occur.

For axially symmetric distributions the interactions [43] can be written in simpler form using [17], as was the case for the electrostatic interaction.

The dipole–dipole term of [43] can be transformed into (Gray 1976)

\[ V_{11} = \frac{1}{R^3} \left[ m_i \cdot m_j - \frac{3(m_i \cdot R)(m_j \cdot R)}{R^2} \right] \]

the familiar Cartesian expression.

5. Spin Contributions to the Multipolar Interactions

Particle \( i \) is now assumed to have an intrinsic magnetic dipole moment \( m_i = (e\hbar/2m_\mu)c \mathbf{s}_i \) associated with its spin \( \mathbf{s}_i \). The contribution to the magnetic vector potential at \( R \) due to the spin magnetic dipole moment of particle \( i \) is (cf. [11])

\[ A'_i(R) = -\nabla_i \times \frac{m_i}{|R - r_i|} \]

Also the interaction energy of the spin magnetic dipole with an external potential \( A(r) \) is (cf. [31])

\[ V'_{ij} = -m_i \cdot B(r_j) = -m_i \cdot \nabla_i \times A(r_j) \]

Substituting the expansion (Jackson 1962; Gray 1976) for \( |R - r_i|^{-1} \) in [45], and [26] in [46], we find that the results [11] and [31] of Sects. 2 and 3 respectively remain valid if the spin contribution

\[ M_{lm}' = \sum l (e\hbar/2m_\mu)c \mathbf{s}_i \cdot \nabla_r \theta^l_l C_{lm}(\omega) \]

is added to the orbital multipole moment [13]. We see from [47] that the spin magnetic dipole moment in general generates additional moments of all orders when displaced from the origin. We also note from [47] and \( \nabla_r = 1 \) that the dipole term is independent of origin, i.e.

\[ M_{1l} = \sum r_i \]

It now follows that [43] is generally valid if \( M_{lm} \) is interpreted as the sum of orbital and spin components. In the general magnetic interaction between two molecules (1) and (2) there are therefore terms of the types \( M_{lm}(1) M_{ln}(2), M_{lm}(1) M_{ln}(2), M_{lm}(1) M_{ln}(2) \), and \( M_{lm}(1) M_{ln}(2) \), corresponding to the intramolecular orbit–orbit, spin–other orbit, and spin–spin contributions as normally defined (Slater 1960).

6. Conclusion

Elementary methods have been used to derive [22], [31], [33], and [43] which are of fundamental importance in the multipole description of magnetostatics.

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Appendix: The Darwin Correction

Although the contribution $A_i(R)$ of the charge $e_i$ to the vector potential at $R$ is not divergence-free, the total vector potential

$$A(R) = \sum_i A_i(R) = \sum_i \frac{e_i v_i}{|R - r_i|c}$$

has no divergence (Van Vleck 1932). Therefore the Darwin correction to $A_i(R)$

$$\frac{1}{2c} \sum_i \left[ e_i v_i \frac{(R - r_i)(R - r_i)}{|R - r_i|^3} - \frac{e_i v_i}{|R - r_i|} \right]$$

which arises from consideration of retardation effects (Darwin 1920; Slater 1960), and which also ensures that the individual $A_i(R)$ are divergence-free (Itoh 1965), must vanish for a stationary distribution, since the vector potential with gauge condition $\nabla \cdot A = 0$ is unique.

The fact that [49] vanishes can also be seen by direct calculation. To simplify the proof we go over to the continuous representation. Using $\nabla \cdot J = 0$ we see that the first term in [49] is equal to

$$\frac{1}{2c} \int dr \frac{J(r) \cdot (R - r)(R - r)}{|R - r|^3} = \frac{1}{2c} \int dr \left\{ \nabla_r \left[ \frac{J(r)(R - r)}{|R - r|} \right] + \frac{J(r)}{|R - r|} \right\}$$

where the usual convention $\nabla \cdot (AB) = \nabla_a (A_a B)$ is employed. The first term on the right hand side of [50] vanishes when Gauss’s theorem is applied to it. This leaves the second term which corresponds to the second term in [49]. Thus, the two terms in [49] cancel if the current distribution is stationary.

The Darwin correction

$$\frac{1}{2c} \sum_{i<j} e_i e_j \left[ \frac{v_i \cdot v_j}{r_{ij}^3} - \frac{v_i \cdot v_j}{r_{ij}} \right]$$

to the interaction energy between two current distributions is seen to vanish in the same way.
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