Irreducible tensor form for the AME coupling

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Abstract

Using the multipole expansion of electromagnetic (EM) field, we present the angular magnetoelectric (AME) coupling in irreducible tensor form. We evaluate the matrix elements when the radiation source is described by electronic transitions in atomic systems. The results indicate that the energy corrections increase for short wavelengths and large charge number.

Keywords: AME coupling, irreducible tensor form, multipole expansion

1. Introduction

In consequence of their results on the time-dependent Foldy–Wouthuysen transformation [1], Mondal et al show [2] that the AME coupling represents the $O(c^{-2})$ correction to the Dirac operator for an electron in a potential $V$ and interacting with the EM field, which is characterized by the 4-component vector potential $A' = (\Phi, A)$ (for an alternative derivation we refer to [3]):

$$H_{AME} = \frac{1}{2c^2} (H^{\text{in}}_{AME} + H^{\text{ex}}_{AME})$$

(1.1)

where

$$H^{\text{in}}_{AME} = S \cdot (E \times A), \quad E = -\nabla V,$$

(1.2a)

and $S$ is the spin operator. Throughout we use atomic units unless explicitly stated otherwise. One calls $H^{\text{in}}_{AME}$ (resp. $H^{\text{ex}}_{AME}$) an intrinsic (resp. induced) part of AME coupling. In the Coulomb gauge fixing, $H^{\text{ex}}_{AME}$ serves as the source for obtaining the “hidden energy” that couples the EM angular momentum density with magnetic moments [4].

It is our purpose here to investigate the input of $H_{AME}$ into the atomic energy levels provided that the radiation source is defined by electronic transitions. The exploration of the standard multipole expansion of EM field [5] allows us to represent $H_{AME}$ as the summable series (in the sense of distributions) of irreducible tensor operators, while the information about the radiation source is contained separately, in the coefficients of expansion (amplitudes).

As one could expect, the contribution of the AME coupling to the total energy should be small enough. For example, we show that the matrix elements of the intrinsic part are of order $O(\omega^2 Z^2 c^{-3})$ for the E1 transition, while the matrix elements of the induced part are of order $O(\omega^2 Z^2 c^{-6})$ for the same type of radiation; here $\omega$ is the transition energy. However, $\omega$ usually increases as the charge number $Z$ becomes large, which results in larger energy corrections.

By (1.2), $H^{\text{in}}_{AME}$ is time-dependent, while $H^{\text{ex}}_{AME}$ is time-independent. The latter is in agreement with [2], where the plane EM wave expansion is used for explaining the inverse Faraday effect. In addition, we show that $H^{\text{ex}}_{AME}$ can be split in two separate parts. One part is traditional in the sense that it does not vanish if the multipole moment of order $l \in \mathbb{N}$ is nonzero for at least one fixed $l$, while the second one is more “exotic” in the sense that it can be nonzero only if the multipole moment is nonzero for at least two different $l$. The latter case arises when, for example, one considers electronic satellite transitions produced by electron capture and subsequent radiative decay [6–10].

In Sec. 2 we express (1.2) in irreducible tensor form. We work in the Coulomb gauge fixing and we use the standard technique of angular momentum theory [11–14] (including the notation and the phase system used therein). We discuss the matrix elements in particular cases in Sec. 3.

2. Tensor operators

2.1. Amplitudes

Let the radiation of energy $\omega = \nu c = E_{\alpha'J'} - E_{\alpha J}$ be emitted by the electron going from the state $|\alpha J M\rangle$ to the (lower) state $|\alpha' J'M'\rangle$; $\alpha$ and $\alpha'$ denote additional quantum numbers if necessary. When $\nu \ll 1$, the amplitudes for the radiation of order $(l, m), l \in \mathbb{N}, m \in \{-l, \ldots, l\}$, are approximated by [5]

$$a^{\rho}_{l,m} = \delta_{\rho M} a^{\alpha}_{l,M'}, \quad \rho = M - M'. \quad (2.1)$$

Here the superscript denotes both $E$ (electric type) and $M$ (magnetic type), and

$$a^{E}_{l,M} = \lambda_{l,M} Q_{l,M}, \quad a^{M}_{l,M} = -\lambda_{l,M} M_{l,M}. \quad (2.2)$$

The multiplier

$$\lambda_{l,M} = (\sqrt{2J + 1})^{l+1} \frac{\Gamma(2l+1)\sqrt{4\pi(2l+1)}}{\sqrt{2J + 1}}$$

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\[ J' \left\| M' \right\| J \left\| M \right\| \right], \quad K_i = -\sqrt{1 + 1/l}. \tag{2.3} \]

The number \( Q_\omega \) (resp. \( M_\rho \)) is the reduced matrix element of the electric (resp. magnetic) multipole moment \( Q' \) (resp. \( M' \)):

\[ Q_{\omega l} = (\alpha' J' \left\| Q' \right\| \alpha J), \quad M_{\rho l} = (\alpha' J' \left\| M' \right\| \alpha J). \tag{2.4} \]

When the magnetization is ignored, we have

\[ Q' = -i' C, \quad M' = -\frac{\sqrt{l(2l-1)}}{c(l+1)} J^{-1}[C^{-1} \times L^1]' \tag{2.5} \]

Otherwise: \( Q' \) is replaced by \( Q' + O(\nu/c) \), hence we omit the \( O(\nu/c) \) correction since we already have the small \( \nu'^2 \) in (2.3); \( M' \) is replaced by \( M' + M'^1 \), where

\[ M'^1 = -\frac{1}{c} \frac{\sqrt{l(2l-1)}}{c(l+1)} J^{-1}[C^{-1} \times S^1]' \tag{2.6} \]

In the examples to be followed, we assume \( M' + M'^1 \) when we write \( M' \); see also [11, Secs. 4 and 25].

### 2.2. Intrinsic part

As in [2], we take the real part of the external electric field \( E^r \). Applying the well-known angular momentum technique, we deduce from [5, Appendix B.2] the following form for the intrinsic part \( H^{\text{AM}}_{\text{AME}} \equiv (H^{\text{AM}}_{\text{AME}})^E \) of electric type

\[ (H^{\text{AM}}_{\text{AME}})^E_{r l} = \sum_{l=0}^{l} \delta^E_{l l m} C_{l l m} J^{-1} \left\{ C_{l l m} \right\} \tag{2.7a} \]

where the rank-\( l \) irreducible tensor operator

\[ (H^{\text{AM}}_{\text{AME}})^E_{l l} = \frac{-i^{-l} V(r)}{2 \omega \sqrt{l(l+1)}} \left\{ C_{l l} \times S^1 \right\} \tag{2.7b} \]

\[ \cdot [l(l+1) J_{l l} - l J_{l l}] \]

\((j)\) is the spherical Bessel function and the amplitude

\[ \delta^E_{l l m}(t) = \frac{1}{2} (\epsilon_{l l m} E_{l l m} (l (l+1)) + (l-m) \epsilon_{l l m} E_{l l m} (l (l+1) + m)) \tag{2.7c} \]

Here \( \sigma_j = \arg \Gamma(l+1+i\eta) \) is the Coulomb phase shift, \( \eta = -Z/v \) is the Sommerfeld parameter.

Likewise, the intrinsic part \( H^{\text{AM}}_{\text{AME}} \equiv (H^{\text{AM}}_{\text{AME}})^M \) of magnetic type is written in the form (2.7a), but with the superscript \( E \) replaced by the superscript \( M \), and with the amplitude replaced by

\[ \beta^M_{l l m}(t) = \frac{1}{2} (\epsilon_{l l m} E_{l l m} (l (l+1)) + (l-m) \epsilon_{l l m} E_{l l m} (l (l+1) + m)) \tag{2.8a} \]

The corresponding rank-\( l \) irreducible tensor operator

\[ (H^{\text{AM}}_{\text{AME}})^M_{l l} = \frac{i^{-l-1} V(r)}{2 \omega \sqrt{l(l+1)}} \left\{ C_{l l} \times S^1 \right\} \tag{2.8b} \]

\[ \cdot \left[ (l(l+1)(2l+1)) C_{l l} \times S^1 \right] - \sqrt{l(l+1)(2l+1)(2l+3)} \epsilon_{l l m} E_{l l m} \epsilon_{l l m} E_{l l m} \]

\[ \cdot \left[ [(l+1)(l+2)(2l+3)] J_{l l} - l J_{l l}] \right] \]

In [2] the authors put \( A = B \times x/2 \) for almost every \( x \in \mathbb{R}^3 \). In this case \( \text{div} A^M = 0 \) but \( \text{div} A^E \neq 0 \); for \( A = A^M \) of magnetic type, \( j_l(\nu) \) in (2.8b) is replaced by \( [(l+1) j_{l+1}(\nu) - \nu j_{l+1}(\nu)]/[2] \).

From the point of view of energy levels, the treatment of \( H^{\text{AME}}_{\text{AME}}(2c^2) \), when considered as the \( O(c^{-2}) \) correction to the Pauli operator for an electron in a potential \( V \), is subtle in that it is time-dependent. We refer to [15–17], where the eigenvalue problem for the time-dependent Pauli equation is studied in detail.

### 2.3. Induced part

Unlike the intrinsic part of AME coupling, the induced part contains the products of the time-dependent amplitudes \( a^\text{AM}_{l l m}(t) \) and \( \beta^\text{AM}_{l l m}(t) \); here \( \alpha^\text{AM}_{l l m}(t) \) (resp. \( \beta^\text{AM}_{l l m}(t) \)) is defined by (2.7c) (resp. (2.8a)), but with the superscript \( E \) (resp. \( M \)) replaced by the superscript \( E \) (resp. \( M \)). However, using the symmetry properties of the products and interchanging the summation indices \( l \) and \( J \) we find that the induced part of AME coupling is actually time-independent. As a result, \( H^{\text{AM}}_{\text{AME}} \equiv (H^{\text{AM}}_{\text{AME}})^E \) splits into two parts:

\[ (H^{\text{AME}}_{\text{AME}})^E = (H^{\text{AME}}_{\text{AME}})^{E'} + (H^{\text{AME}}_{\text{AME}})^{E''}. \tag{2.9} \]

For the radiation of electric type we have

\[ (H^{\text{AME}}_{\text{AME}})^E_{l l} = \frac{-i l}{2} \sum_{J=0} A_{l l J} \left( \begin{array}{c} 1 \ \ l \ \ J \\ J \ \ -J \ \ 0 \end{array} \right) \] \[ \left( \begin{array}{ccc} 1 & l & J \\ J & -J & 0 \end{array} \right) \] \[ \left( \begin{array}{ccc} 1 & l & J \\ J & -J & 0 \end{array} \right) \] \[ \left( \begin{array}{ccc} 1 & l & J \\ J & -J & 0 \end{array} \right) \]

(2.10a)

(2.10b)

(2.11)

and \( (H^{\text{AME}}_{\text{AME}})^E_{l l J} \) is the 0th component of the rank-J \((l + l' \leq J \leq l + l')\) tensor operator

\[ (H^{\text{AME}}_{\text{AME}})^E_{l l J} = \frac{\sqrt{j_{l+1} \Gamma l}}{2 \pi} \sum_{l' l''} A_{l l J} \left( \begin{array}{c} 1 \ \ l' \ \ l'' \\ J \ \ -J \ \ 0 \end{array} \right) \]

\[ \left( \begin{array}{ccc} 1 & l' & l'' \\ J & -J & 0 \end{array} \right) \]

\[ \left( \begin{array}{ccc} 1 & l' & l'' \\ J & -J & 0 \end{array} \right) \]

\[ \left( \begin{array}{ccc} 1 & l' & l'' \\ J & -J & 0 \end{array} \right) \]

\[ \left( \begin{array}{ccc} 1 & l' & l'' \\ J & -J & 0 \end{array} \right) \]

(2.11a)

(2.11b)
The integers $k$ are such that $k + l + l'$ is even and at least one of the following four conditions holds:

- $\max[|J - 1|, |l - l'|] \leq k \leq \min(J + 1, l + l' + 2)$
- $\max[|J - 1|, |l - l'| + 2] \leq k \leq \min(J + 1, l + l')$

Notice that $J$ in (2.10a) is necessarily odd, because

$$
(H^e_{\text{AME}})^{E\rho}_{\nu l j} = (-1)^{j + l + l'}(H^e_{\text{AME}})^{E\rho}_{\nu l j}.
$$

(2.13)

It follows from above that:

- $(H^e_{\text{AME}})^{E\rho}_{\nu l j} = 0$ for $\rho = 0$ (i.e. $M = M'$).
- $(H^e_{\text{AME}})^{E\rho}_{\nu l j}$ is nonzero if $Q_{\rho l}$ is nonzero for at least two different values of $l$.

For the radiation of magnetic type, the terms $(H^e_{\text{AME}})^{M\nu}_{\rho l j}$ and $(H^e_{\text{AME}})^{M\nu\prime}_{\rho l j}$ are given by (2.10a) and (2.10b), respectively, but with the superscript $E$ replaced by the superscript $M$, the phase $(-1)^{\rho l}$ replaced by $(-1)^{\rho l'}$, and with the corresponding rank-$J$ tensor operator

$$
(H^e_{\text{AME}})^{M\nu}_{\rho l j} = \sqrt{\frac{(2l + 1)(2l' + 1)}{2\pi a_{\nu l j}}}
$$

\[ \cdot j_l(vr) j_{l'}(vr) \sum_k k^2 \sqrt{k + 1} \left[ C^k \times S^1 \right]^j \]

\[ \cdot \left[ \begin{array}{c} l' \ l \ k' \ l \ j \ k \end{array} \right] \left[ \begin{array}{c} l' \ l \ k \ j \ k \end{array} \right].
$$

(2.14)

The integers $k$ are such that $k + l + l'$ is even and it holds $\max[|J - 1|, |l - l'|] \leq k \leq \min(J + 1, l + l')$. For $A = B \times \pi/2$ of magnetic type, $j_l(vr)$ in (2.14) is replaced by $[(l + 2) j_l(vr) - vr j_{l'}(vr)]/2$.

Notice that $(H^e_{\text{AME}})^{M\nu}_{\rho l j}$ satisfies the symmetry property analogous to (2.13). In addition:

- Since the 9 $j$-symbol $\left[ \begin{array}{c} l' \ l \ k \end{array} \right] = 0$ for $k = J$ (see e.g. [13, Eq. (31.10)]), and since $k + l + l'$ is even, the total $(H^e_{\text{AME}})^{M\nu}_{\rho l j} = 0$ for $\rho = 0$.
- $(H^e_{\text{AME}})^{M\nu\prime}_{\rho l j}$ is nonzero if $M_{\rho l}$ is nonzero for at least two different values of $l$ (provided $\rho \neq 0$).

Although it should be obvious, we would like to emphasize that $J$ in Sec. 2.3 is not the same as $J$ in (2.1).
According to [19, Tab. 3], $\omega = \omega(Z)$ is an increasing function; for example, $\omega$ is around 597402 (cm$^{-1}$) in Ca XVI and around 847413 (cm$^{-1}$) in Zn XXVI. This indicates that the contribution of the intrinsic part of AME coupling to the total energy increases when $Z$ becomes large.

For $M = -M' = 1/2$ (i.e. $\rho = 1$), the first nonzero matrix elements in the induced case are given by (note that the tensor operator in (2.10b) vanishes)

\[
\langle 1s, \pm 1/2 | ^{E'\nu}_{\mathrm{AME}} | 1s, \pm 1/2 \rangle = \pm \frac{1}{12} \nu^5 + O(\nu^7), \quad (3.3)
\]

\[
\langle 1s, \pm 1/2 | ^{E''\nu}_{\mathrm{AME}} | 1s, \pm 1/2 \rangle = \pm \frac{1}{184320} \nu^{11} + O(\nu^{13}).
\]

We plot $\nu^5/(12\nu^2Z^2)$ in Fig. 1, from where we conclude that the increasing $\nu^5$ tends to dominate over $Z^{-2}$.

Table 1: The value of $(1s, 1/2 | ^{E'\nu}_{\mathrm{AME}} | 1s, 1/2)$ for a given electronic transition in Zn XXVI. The numbers are in 10$^{-18}$ cm$^{-1}$.

| Transition          | Matrix element |
|---------------------|----------------|
| $2s^2p^2 3s^1/2 - 2s^2p^2 3p^1/2$ | 16.3051 |
| $2s^2p^2 3s^1/2 - 2s^2p^2 3p^1/2$ | 3.2119 |
| $2s^2p^2 3p^1/2 - 2s^2p^2 3p^1/2$ | 35.9883 |
| $2s^2p^2 3p^3/2 - 2s^2p^2 3p^3/2$ | 40.8814 |
| $2s^2p^2 23/2 - 2s^2p^2 23/2$ | 4.4750 |
| $2s^2p^2 23/2 - 2s^2p^2 23/2$ | 2.0247 |

The matrix elements for some $E_1$ transitions are listed in Tab. 1. In comparison, for $E2$ (M1) transition $2s^2p^2 3p^3/2 - 2s^2p^2 3p^1/2$, the matrix element of $^E\nu_{\mathrm{AME}}$ is of order $O(\nu^7Z^{-6}c^{-3})$ ($O(\nu^{11}Z^{-2}c^{-3}$)).

3.2. Electronic satellite transitions

So far we have considered transitions of a particular order $l$. Now we give examples for the term in (2.10b).

Let the radiation source be the decay of type

\[ n_1 | l^1 \rangle + n_2 | l^2 \rangle + n_3 | l^3 \rangle + n_4 | l^4 \rangle - n_1 | l^{1+2} \rangle - n_2 | l^{2+3} \rangle - n_3 | l^{3+4} \rangle - n_4 | l^{4+1} \rangle \]

of wavelength $\gg 21$ (Å), i.e. $\nu \ll 1$. The radiation of this type produces satellite lines considered e.g. in [7]. For the transition in Yb-like W [7, Tab. 6]

\[
4f^{14}5p^5 6s 6d^3 \Gamma_6 \rightarrow 4f^{14}5p^5 6s 6d^3 \Gamma_5
\]

of wavelength 410.6 (Å) (that is, $\nu \approx 0.050928$) we get that $Q_{\ell}$ is nonzero for $l = 1, 3, 5$, where

\[
Q_{\ell 1} = \sqrt{\frac{39}{7}} \langle r^5 \rangle_{sd, sf}, \quad Q_{\ell 3} = \frac{3}{5} \sqrt{\frac{26}{3}} \langle r^3 \rangle_{sd, sf},
\]

and where the radial integrals

\[
\langle r^5 \rangle_{sd, sf} = -\frac{1}{65} 2^{-7} \nu^{-2} Z^{-1} \cdot (l + 2)(16 + l(l + 3))\Gamma(l + 8).
\]

Let $M = 1$ and $M' = 0$ so that $\rho = 1$. The matrix elements of $(^E\nu_{\mathrm{AME}})_{\ell}$ are of order $O(\nu^7 Z^{-6} c^{-3})$; e.g.

\[
\langle 1s, 1/2 | ^{E'\nu}_{\mathrm{AME}} | 3d, 1/2 \rangle = (3d_{-1, 1/2} | ^{E'\nu}_{\mathrm{AME}} | 1s, 1/2) \quad (3.4a)
\]

\[
= \frac{3825 \sqrt{5} 1^9}{2464} \sin (\sigma_1 - \sigma_3) + O(\nu^{11}) \cdot (3.4b)
\]

Therefore, the eigenvalues of the (Hermitian) matrix are of order $10^{-24} \sin(\sigma_1 - \sigma_3) (\text{cm}^{-1})$. To compare with, for $M' = -1$ the above matrix element is of order $O(\nu^{17} Z^{-13} c^{-3}) \sin(\sigma_1 - \sigma_3)$. There are transitions (e.g. when $l = 2, 4$) for which the matrix elements are proportional to $\cos(\sigma_1 - \sigma_3)$; hence theoretically the elements are nonzero even if one puts the Coulomb phase shift to 0. On the other hand, the above example suggests that the contribution of the present part of AME coupling is practically negligible.

The major cause of the small values of matrix elements is the small $\nu$. To make $\nu$ large the satellite lines are a good example, for their wavelengths can be less than 21 (Å) [6, 8]. In order to use the transitions with $\nu \gtrsim 1$ we need to modify the definition of amplitudes. Here we only consider the radiation of electric type. With the help of [5, Appendix B, Eq. (4.10)] we find that the multiplier $A_{\ell \omega}$ in (2.3) is of the form

\[
A_{\ell \omega} = (-1)^{l-j'} \frac{i^{j'2} \sqrt{l(l+1)}}{2^{j'1+1}} \sqrt{4\pi(2l+1)} \left[ J' \frac{l}{M'} \rho M \right] \quad (3.5)
\]

and the multipole moment is given by

\[
Q' = [\langle r_j \rangle (\nu r)] C' + O(\nu/c) \quad (3.6)
\]

where the prime denotes the derivative with respect to $r$. Since we still have $\omega \ll c^2$, we do not give an explicit representation for the term $O(\nu/c)$, which we omit in what follows.

Subsequently, the radial integral (3.1a) is replaced by

\[
R_{\ell \omega}(\alpha_1, \alpha_2) = \int_0^\infty R_{\alpha_1}(\nu) [\langle r_j \rangle (\nu r)] R_{\alpha_2}(\nu) r^2 \: dr. \quad (3.7)
\]
One verifies that \( a_r^E \) in (2.2), with \( \lambda \) and \( Q \) as in (3.5) and (3.6), approaches \( a_r^E \) with \( \lambda_o \) and \( Q^o \) as in (2.3) and (2.5), as \( \nu \rightarrow 0 \).

As an example, let us consider the Rydberg transition in Ag-like W\(^{27+}\) ion [8, Tab.6]

\[
4d^4f(LS)\,9f^2G_{9/2} - 4d^{10}4f^2F_{7/2}
\]

of wavelength 13.2 (\( \angstrom \)), i.e. \( \nu \approx 1.58417 \). Let \( (LS) = \nu \); then

\[
Q_{r1} = \frac{3}{7} \sqrt{\frac{19}{2}} R_{r1}(4d, 9f),
\]

\[
Q_{r3} = \frac{1}{7} \sqrt{\frac{418}{21}} R_{r3}(4d, 9f),
\]

\[
Q_{r5} = \frac{1}{7} \sqrt{\frac{247}{66}} R_{r5}(4d, 9f).
\]

For \( M = -M^* = 1/2 \), the matrix element (3.4) now takes the value (in cm\(^{-1}\))

\[
\langle 1s, \, 1/2 | (H_{AME}^{ex} \cdot \vec{E}) | 3d_\nu \, , \, 1/2 \rangle \\
\approx -1.47049 \cdot 10^{-14} \, i \sin(\sigma_1 - \sigma_3)
\]

\[
+ 8.04766 \cdot 10^{-27} \, i \sin(\sigma_3 - \sigma_5)
\]

while the matrix element (3.3) is given by

\[
\langle 1s, \, \pm 1/2 | (H_{AME}^{ex} \cdot \vec{E}) | 1s, \, \pm 1/2 \rangle \approx \pm 9.33947 \cdot 10^{-7}.
\]

Since the expansion in powers of \( \nu \) no longer makes sense, and the analytical representation of the matrix elements is rather complicated and veiled, we show only numerical values. As expected, the eigenvalues of the corresponding matrices are much larger compared to those for the previous transitions. Yet the values are much smaller compared to the classical \( O(\nu^2) \) corrections. On the other hand, the matrix element (3.2), which now equals \( \pm 10.9953 \, \sin(\alpha + \sigma_1) \, (\text{cm}^{-1}) \), allows one to expect a sizeable contribution to energy levels.

### 4. Concluding remarks

We present the AME coupling term as the summable series of irreducible tensor operators so that the whole information about the radiation source is contained in the coefficients of expansion. The form is convenient both for applying directly the Wigner–Eckart theorem and for changing the radiation source, depending on ones interest, without affecting the structure of tensor operator.

We give a rough estimation of the order of magnitude of the corresponding matrix elements by considering electronic transitions in atomic systems. The examples indicate that the most valuable contribution to energy levels comes from the time-dependent intrinsic part of the AME coupling, provided that the radiation source is described by electronic transitions at short wavelengths and that the charge number is large enough.

A rigorous investigation of the eigenvalue problem for the time-dependent Pauli operator [15] perturbed by the time-dependent part of AME coupling still needs to be done. From the analytical point of view it would also be of interest to investigate the change of the AME coupling when modifying the vector potential by using gauge transformations.

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