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1987 J. Phys. A: Math. Gen. 20 825

(http://iopscience.iop.org/0305-4470/20/4/018)

J. Phys. A: Math. Gen. **20** (1987) 825-837. Printed in the UK

Differentiation of spherical tensors: application to the determination of polarisability tensors†

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Received 4 December 1985, in final form 23 June 1986

Abstract. The differentiation of tensors in spherical coordinates is analysed for two distinct cases: when the result is a tensor of a rank higher, or lower, than the spherical tensor subjected to differentiation. General analytical expressions are obtained leading to equations applicable to particular problems of the theory of light scattering and absorption.

The relationships obtained are used for defining spherical tensors of polarisability a_{ij}, hyperpolarisability B_{ijk} and dipole-quadrupole polarisability $A_{i,jk}$ for isolated molecules as well as the tensor of interaction-induced polarisability of a molecular sample taking into account intermolecular interactions in the approximation of the dipole-induced dipole (DID) model.

1. Introduction

Certain tensorial quantities, for example the directional force of an oscillator, the stress tensor, electric multipole moments, the polarisabilities of molecules, etc, are defined in cartesian space by the use of derivatives of the appropriate tensors with respect to the others (Kielich 1981, Applequist 1984). The tensors thus defined are then transformed directly or by the step-by-step coupling method to a system of spherical coordinates (Coope 1970, Stone 1975, 1976, Normand and Raynal 1982). Often, the tensor obtained by differentiation in cartesian coordinates is of a structure so highly complicated as to render its transformation to spherical coordinates extremely tedious. It is then more convenient to transform the tensor prior to differentiation (its structure is then quite simple) into spherical coordinates first and then to perform the operation of differentiation in spherical coordinates.

In the present paper, we shall be dealing with the differentiation of spherical tensors. In § 3 we analyse the problem of differentiating a spherical tensor as the result of which one obtains a tensor of a rank higher than that of the initial tensor. This is the case, for example, when it comes to determine a generalised electric multipole polarisability. With this aim, we apply the expression proposed by Kielich (1965a, b, 1966) for the induced molecular multipole moment of order n:

$$\boldsymbol{m}^{\{n\}}=\frac{1}{n!}\sum_{n_1=1}^{\infty}\dots\sum_{n_s=1}^{\infty}\frac{2^{n_1+\dots+n_s}n_1!\dots n_s!}{(2n_1)!\dots(2n_s)!}\boldsymbol{a}^{\{n+n_1+\dots+n_s\}}\langle n_1+\dots+n_s\rangle\boldsymbol{E}^{\{n_1\}}\boldsymbol{E}^{\{n_2\}}\dots\boldsymbol{E}^{\{n_s\}}$$

† This work was carried out under research project CPBP 01.12.5.6.

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where $E^{\{n_i\}}$ is the amplitude of the n_i-fold gradient of the external electric field strength and $\langle n_1+\ldots+n_s\rangle$ stands for $(n_1+\ldots+n_s)$-fold contraction. The polarisability tensor of rank $(n+n_1+\ldots+n_s)$ is obtained on performing the following operation of differentiation (Applequist 1984):

$$a^{\{n+n_1+\ldots+n_s\}}=\partial^s m^{\{n\}}/\partial E^{\{n_1\}}\ldots\partial E^{\{n_s\}}.$$

We shall be having recourse to particular cases of these two equations in the course of the present study.

In § 4, we consider the case when the spherical tensor corresponds to a cartesian polyade (affinor) and is to be differentiated with respect to one of the elements of the polyade. One then arrives at a spherical tensor of a rank lower than that of the tensor subjected to differentiation. This is the situation we deal with when differentiating a polyade of the nth rank $A^{\{n\}}=A^{\{1\}}\ldots n$ times $\ldots A^{\{1\}}$, formed from the vector $A^{\{1\}}$ with respect to an arbitrary component of the latter.

Section 5 is devoted to the determination of spherical tensors of molecular polarisability by the technique of differentiation in a system of spherical coordinates. Our calculations lead to the dipolar polarisability tensor for an isolated molecule and to the tensor of interaction-induced polarisability in the DID approximation. Moreover, a relation is established between the spherical hyperpolarisability tensor and the second derivative of the dipole moment of the molecule with respect to variations of the external electric field. The last example of the application of the formulae derived in this work is related to the determination of the spherical tensor of dipole–quadrupole molecular polarisability, where we have drawn attention to the problem of symmetrisation of the tensors obtained as the result of differentiation.

2. The fundamental equations

Let us consider a system of reference defined by the components e_i $(i=x, y, z)$ of the unit vector e. We use these components to construct a circular system $\mathbb{C}$ determined by the unit vector $e^{(1)}$ with the following components, in the phase convention of Fano and Racah (1959)

$$e^{(1)}_{\pm 1}=\mp\frac{\mathrm{i}}{\sqrt{2}}(e_x\pm \mathrm{i}e_y) \qquad e^{(1)}_0=\mathrm{i}e_z. \tag{1}$$

These components (1) form the self-conjugate and normalised basis of the circular system

$$e^{(1)}_\alpha e^{[1]}_\beta=\delta_{\alpha\beta} \tag{2}$$

where $e^{[1]}$ is a vector of the contravariant basis (Varshalovich *et al* 1975, Ożgo 1975).

The inner product of the first kind (scalar product) of two vectors A and B defined in the system $\mathbb{C}$ is

$$U_1=A\cdot B=\sum_\alpha A^{(1)}_\alpha B^{[1]}_\alpha=\sum_\alpha(-1)^{\alpha+1}A^{(1)}_\alpha B^{(1)}_{-\alpha}. \tag{3}$$

This product, being invariant with respect to transformations from one system of reference to another, can serve as a method of determining the shape of tensors in a selected system.

We now proceed to calculate the scalar product of the cartesian tensor $A_{i_1\ldots i_n}$, which transforms according to the three-dimensional group of rotations, with the unit vector in n-dimensional space

$$U_n = A_{i_1\ldots i_n} \prod_{j=1}^{n} e_{i_j} = \sum_{\alpha_1\ldots\alpha_n} \prod_{i=1}^{n} A^{(1)}_{\alpha_i} e^{[1]}_{\alpha_1} \ldots e^{[1]}_{\alpha_n} \tag{4}$$

where we had recourse n times to the relation (3) and took into account that $A_{i_1\ldots i_n} = A_{i_1}\ldots A_{i_n}$. Equation (4) contains the n-fold irreducible tensorial product $\Pi\, \boldsymbol{A}^{(1)}$ (coupling of tensors) which, for two spherical tensors of the ranks k and l, is defined as follows (Varshalovich *et al* 1975):

$$C^{(kln)}_{\nu} = (\boldsymbol{A}^{(k)} \otimes \boldsymbol{B}^{(l)})^{(n)}_{\nu} = \sum_{\lambda\mu} (-1)^{k-l+\nu} [n] \begin{pmatrix} k & l & n \\ \lambda & \mu & -\nu \end{pmatrix} A^{(k)}_{\lambda} B^{(l)}_{\mu}. \tag{5a}$$

On inversion of (5a), we obtain

$$A^{(k)}_{\lambda} B^{(l)}_{\mu} = \sum_{n} (-1)^{k-l+\nu} [n] \begin{pmatrix} k & l & n \\ \lambda & \mu & -\nu \end{pmatrix} (\boldsymbol{A}^{(k)} \otimes \boldsymbol{B}^{(l)})^{(n)}_{\nu}. \tag{5b}$$

The subscript $\lambda(\mu)$ labels the $(2k+1)$ components of the tensor $\boldsymbol{A}^{(k)}$ (the $(2l+1)$ components of $\boldsymbol{B}^{(l)}$), whereas $[ab\ldots f] = [(2a+1)(2b+1)\ldots(2f+1)]^{1/2}$ and $\left(\begin{smallmatrix} k & l & n \\ \lambda & \mu & \nu \end{smallmatrix}\right)$ is a $3j$ coefficient.

In conformity with the principle of tensor coupling, (5b) yields

$$\prod_{i=1}^{n} A^{(1)}_{\alpha_i} = \sum_{d_1\ldots d_n} \prod_{i=1}^{n-1} (-1)^{d_i-1+\nu_{i+1}} [d_{i+1}] \begin{pmatrix} d_i & 1 & d_{i+1} \\ \nu_i & \alpha_{i+1} & -\nu_{i+1} \end{pmatrix} A^{(d_1\ldots d_n)}_{\nu_n}. \tag{6}$$

Thus, the spherical tensor $\boldsymbol{A}^{(d_1\ldots d_n)}$ of rank d_n has arisen as the result of $(n-1)$ vector couplings within the nth rank polyade:

$$A^{(d_1\ldots d_n)}_{\nu_n} = \bigotimes_{i=1}^{n} A^{(1)}_{\alpha_i} \tag{7}$$

where $\nu_n = \Sigma\, \alpha_i$. We use the product of basis vectors of (4) to construct an irreducible basis of nth order in accordance with (5a, b):

$$e^{(d_1\ldots d_n)}_{\nu_n} = \sum_{\substack{\alpha_2\ldots\alpha_n \\ \nu_1\ldots\nu_{n-1}}} e^{(1)}_{\nu_1} \prod_{i=1}^{n-1} (-1)^{d_i-1+\nu_{i+1}} [d_{i+1}] \begin{pmatrix} d_i & 1 & d_{i+1} \\ \nu_i & \alpha_{i+1} & -\nu_{i+1} \end{pmatrix} e^{(1)}_{\alpha_{i+1}}. \tag{8}$$

The successive steps of coupling, leading to the tensor $\boldsymbol{A}$ (or $\boldsymbol{e}$) in accordance with (7), as determined by $d_1, \ldots, d_{n-1}$, have been written out so as to distinguish different spherical tensors with the same d_n. This was necessary, since knowledge of the transformational properties of a tensor is still insufficient for disclosing the coupling sequence from which it has arisen. It will be remembered that tensors with the same d_n transform identically (Ożgo 1975).

In the preceding formulae, $d_1 = 1$ and $\nu_1 = \alpha_1$. With the basis thus determined, we rewrite (4) and (7) in the form

$$U_n = \sum_{d_1\ldots d_n} \sum_{\nu_n} A^{(d_1\ldots d_n)}_{\nu_n} e^{[d_1\ldots d_n]}_{\nu_n} \tag{9a}$$

$$A^{(d_1\ldots d_n)}_{\nu_n} = A^n e^{(d_1\ldots d_n)}_{\nu_n}. \tag{9b}$$

The basis (8), with respect to (2), is also self-conjugate and normalised in n-dimensional tensorial space:

$$e_{\nu_n}^{(d_1\ldots d_n)} e_{\nu'_n}^{[d'_1\ldots d'_n]} = \delta_{d_1 d'_1} \ldots \delta_{d_n d'_n} \delta_{\nu_n \nu'_n}. \qquad (10)$$

We shall henceforth be dealing with tensors of the type (9b) without entering into the problem of their decomposition into irreducible tensors, or into problems concerning their symmetricity.

3. Differentiation of a spherical tensor A with respect to an unlike tensor B

Let us now consider a cartesian tensor of rank $(m+n)$ obtained by differentiation of a component of an nth rank tensor with respect to another tensor of rank m:

$$L_{i_1\ldots i_n j_1\ldots j_m} = \partial A_{i_1\ldots i_n} / \partial B_{j_1\ldots j_m}. \qquad (11)$$

The above operation enables us to gain information concerning the inner structure of the tensor $\boldsymbol{A}$, i.e. to determine the presence of the tensor $\boldsymbol{B}$ in it and to elucidate how $\boldsymbol{B}$ is connected with the other tensors which, jointly with $\boldsymbol{B}$, form the tensor A. However, differentiation is unable to lead to a decision concerning the nature of the internal connections between the tensors contained in the tensor $\boldsymbol{L}$. This signifies that, for example, contraction

$$L_{i_1\ldots i_n j_1\ldots j_m} := L'_{i_1\ldots i_n j_1\ldots j_m k_1\ldots k_l} C_{k_1\ldots k_l}$$

cannot be detected using (11) alone.

Applying (4) and (6), we transform the left-hand side of (11) to a system of spherical coordinates. This leads to

$$\begin{aligned} U_{n+m} &= L_{i_1\ldots i_n j_1\ldots j_m} \prod_{s=1}^{n} e_{i_s} \prod_{t=1}^{m} e_{j_t} \\ &= \sum_{k_1\ldots k_{n+m}} \sum_{\substack{\alpha_1\ldots\alpha_n \\ \beta_1\ldots\beta_m}} (-1)^{m-\Sigma\beta_t} \prod_{i=1}^{n-1} (-1)^{k_i-1+\sigma_{i+1}} [k_{i+1}] \begin{pmatrix} k_i & 1 & k_{i+1} \\ \sigma_i & \alpha_{i+1} & -\sigma_{i+1} \end{pmatrix} \\ &\quad \times \prod_{j=0}^{m-1} (-1)^{k_{j+n}-1+\sigma_{j+n+1}} [k_{j+n+1}] \begin{pmatrix} k_{j+n} & 1 & k_{j+n+1} \\ \sigma_{j+n} & -\beta_{j+1} & -\sigma_{j+n+1} \end{pmatrix} \\ &\quad \times L_{\sigma_{n+m}}^{(k_1\ldots k_{n+m})} \prod_{s=1}^{n} e_{\alpha_s}^{[1]} \prod_{t=1}^{m} e_{\beta_t}^{(1)}. \end{aligned} \qquad (12)$$

In (12), the basis tensor used in order to represent $\boldsymbol{L}$ in irreducible form has been written as the product of the two bases $\Pi^n e_i$ and $\Pi^m e_j$. This was feasible because the space in which the tensors of the nth and mth rank have been defined is in no direct relationship with the coupling of these tensors. That is to say, coupling does not affect the succession in which the two tensors act (Biedenharn and Louck 1981).

We transform the right-hand side of (11) to spherical coordinates by forming the inner product and step-by-step coupling (cf (7)) of the spherical vectors obtained. We obtain

$$\begin{aligned} U_{n+m} &= \frac{\partial A_{i_1\ldots i_n}}{\partial B_{j_1\ldots j_m}} \prod_{s=1}^{n} e_{i_s} \prod_{t=1}^{m} e_{j_t} \\ &= \partial \left(A_{i_1\ldots i_n} \prod_{s=1}^{n} e_{i_s} \right) \left[\partial \left(B_{j_1\ldots j_m} \prod_{t=1}^{m} e_{j_t} \right) \right]^{-1} \prod_{r=1}^{m} (e_{j_r} e_{j_r}). \end{aligned} \qquad (13a)$$

On contraction of the tensors $\boldsymbol{A}$ and $\boldsymbol{B}$ in the right-hand term of (13*a*) using (4) (which leads to the emergence of scalar expressions in the derivative) and making use of (2), we obtain

$$U_{n+m}=\sum_{l_1\ldots l_n}\sum_{\substack{\varepsilon_1\ldots\varepsilon_m\\ \gamma_1\ldots\gamma_n}}\prod_{i=1}^{n-1}(-1)^{l_i-1+\lambda_{i+1}}[l_{i+1}]\begin{pmatrix} l_i & 1 & l_{i+1}\\ \lambda_i & \gamma_{i+1} & -\lambda_{i+1}\end{pmatrix}$$
$$\times\partial\left(A_{\lambda_n}^{(l_1\ldots l_n)}\prod_{s=1}^{n}e_{\gamma_s}^{[1]}\right)\prod_{t=1}^{m}e_{\varepsilon_t}^{(1)}\left(\sum_{d_1\ldots d_m}\prod_{j=1}^{m-1}(-1)^{d_j-1+\nu_{j+1}}[d_{j+1}]\right.$$
$$\left.\times\begin{pmatrix} d_j & 1 & d_{j+1}\\ \nu_j & \varepsilon_{j+1} & -\nu_{j+1}\end{pmatrix}\partial B_{\nu_m}^{(d_1\ldots d_m)}\right)^{-1}. \qquad (13b)$$

On combining (12) and (13*b*) and on elimination of the *m*-dimensional bases by means of (2), we arrive at

$$\sum_{k_1\ldots k_{n+m}}(-1)^{m-\Sigma\beta_i}\prod_{j=0}^{m-1}(-1)^{k_{j+n}-1+\sigma_{j+n+1}}[k_{j+n+1}]$$
$$\times\begin{pmatrix} k_{j+n} & 1 & k_{j+n+1}\\ \sigma_{j+n} & -\beta_{j+1} & -\sigma_{j+n+1}\end{pmatrix}L_{\sigma_{n+m}}^{(k_1\ldots k_{n+m})}e_{\sigma_n}^{[k_1\ldots k_n]}$$
$$=\sum_{l_1\ldots l_n}\partial A_{\lambda_n}^{(l_1\ldots l_n)}e_{\lambda_n}^{[l_1\ldots l_n]}\left(\sum_{d_1\ldots d_m}\prod_{j=1}^{m-1}(-1)^{d_j-1+\nu_{j+1}}[d_{j+1}]\right.$$
$$\left.\times\begin{pmatrix} d_j & 1 & d_{j+1}\\ \nu_j & \beta_{j+1} & -\nu_{j+1}\end{pmatrix}\partial B_{\nu_m}^{(d_1\ldots d_m)}\right)^{-1} \qquad (14)$$

where the presence of the *n*-dimensional contravariant bases is due to our having applied the rule (8) to the *n*-fold products in (12) and (13*b*). Obviously, these bases can be eliminated from (14) by having recourse to the fact that they are normalised in the meaning of (10). On carrying out the above we immediately obtain the differential of the tensor $\boldsymbol{A}$ in the following form:

$$\partial A_{\sigma_n}^{(k_1\ldots k_n)}=\sum_{\substack{k_{n+1}\ldots k_{n+m}\\ d_1\ldots d_m}}(-1)^{m-\Sigma\beta_i}\prod_{i=1}^{m-1}\prod_{j=0}^{m-1}(-1)^{d_i+k_{j+n}+\nu_{i+1}+\sigma_{j+n+1}}$$
$$\times[d_{i+1}k_{j+n+1}]\begin{pmatrix} d_i & 1 & d_{i+1}\\ \nu_i & \beta_{i+1} & -\nu_{i+1}\end{pmatrix}$$
$$\times\begin{pmatrix} k_{j+n} & 1 & k_{j+n+1}\\ \sigma_{j+n} & -\beta_{j+1} & -\sigma_{j+n+1}\end{pmatrix}L_{\sigma_{n+m}}^{(k_1\ldots k_{n+m})}\partial B_{\nu_m}^{(d_1\ldots d_m)}. \qquad (15a)$$

On summation over $\beta_1,\ldots,\beta_m$ we have

$$\partial A_{\sigma_n}^{(k_1\ldots k_n)}=\sum_{\substack{k_{n+1}\ldots k_{n+m}\\ d_1\ldots d_m}}(-1)^{m+\sigma_n}[k_{n+m}]\begin{pmatrix} k_n & d_m & k_{n+m}\\ \sigma_n & -\nu_m & -\sigma_{n+m}\end{pmatrix}$$
$$\times\prod_{i=1}^{m-1}(-1)^{1+d_i+k_{n+i+1}}[d_{i+1}k_{n+i}]\begin{Bmatrix} d_{i+1} & k_{n+i+1} & k_n\\ k_{n+i} & d_i & 1\end{Bmatrix}$$
$$\times L_{\sigma_{n+m}}^{(k_1\ldots k_{n+m})}\partial B_{\nu_m}^{(d_1\ldots d_m)} \qquad (15b)$$

where we have applied the well known relation (Varshalovich *et al* 1975)

$$\sum_{\alpha\beta\gamma} (-1)^{a+b+c-(\alpha+\beta+\gamma)} \begin{pmatrix} a & d & b \\ \alpha & \delta & -\beta \end{pmatrix} \begin{pmatrix} b & e & c \\ \beta & \varepsilon & -\gamma \end{pmatrix} \begin{pmatrix} c & f & a \\ \gamma & \varphi & -\alpha \end{pmatrix} = \begin{pmatrix} d & e & f \\ -\delta & -\varepsilon & -\varphi \end{pmatrix} \begin{Bmatrix} d & e & f \\ c & a & b \end{Bmatrix}. \tag{16}$$

The symbol $\{\begin{smallmatrix} a & b & c \\ d & e & f \end{smallmatrix}\}$ denotes the 6*j* coefficient. With regard to the fact that, by (9*b*), we have

$$\partial B^{(d_1 \ldots d_m)}_{\nu_m} = \partial B^m e^{(d_1 \ldots d_m)}_{\nu_m}$$

the orthogonality condition (10) valid for different bases $e^{(d_1 \ldots d_m)}$ enables us to write the spherical tensor counterpart of (11) in the following form:

$$\frac{\partial A^{(k_1 \ldots k_n)}_{\sigma_n}}{\partial B^{(l_1 \ldots l_m)}_{\lambda_m}} = \sum_{k_{n+1} \ldots k_{n+m}} (-1)^{m+\sigma_n} [k_{n+m}] \begin{pmatrix} k_n & l_m & k_{n+m} \\ \sigma_n & -\lambda_m & -\sigma_{n+m} \end{pmatrix} \times \prod_{i=1}^{m-1} (-1)^{1+l_i+k_{n+i+1}} [l_{i+1} k_{n+i}] \begin{Bmatrix} k_n & l_{i+1} & k_{n+i+1} \\ 1 & k_{n+i} & l_i \end{Bmatrix} L^{(k_1 \ldots k_{n+m})}_{\sigma_{n+m}}. \tag{17}$$

On the other hand, the tensor $\boldsymbol{L}$ can be regarded as the product of two tensors $a_{i_1 \ldots i_n}$ and $b_{j_1 \ldots j_m}$. In fact, the analytical structure of the right-hand side of (17) resembles that of the spherical tensor representation of the outer product of these two tensors (Stone 1976). It is of essential importance that the tensor $\boldsymbol{L}$—a result of differentiation—permits the unequivocal decision as to which of the tensors $\boldsymbol{A}$ with the same k_n was subjected to differentiation ($\boldsymbol{L}$ is dependent on $k_1 \ldots k_{n-1}$). It is not possible, however, to obtain equally precise information for the tensor $\boldsymbol{B}$. It should be noted that the tensor $\boldsymbol{L}^{(k_1 \ldots k_{n+m})}$, obtained in (17), has the type of symmetry resulting from the coupling scheme (7).

Especially simple is the result of differentiation of the tensor $\boldsymbol{A}$ with respect to variations of the vectorial field ($m = 1$). Here, by (17), we obtain

$$\frac{\partial A^{(k_1 \ldots k_n)}_{\sigma_n}}{\partial B^{(1)}_{\lambda_1}} = \sum_{k_{n+1}} (-1)^{1+\sigma_n} [k_{n+1}] \begin{pmatrix} k_n & 1 & k_{n+1} \\ \sigma_n & -\lambda_1 & -\sigma_{n+1} \end{pmatrix} L^{(k_1 \ldots k_{n+1})}_{\sigma_{n+1}} \tag{18a}$$

whence, by inversion of the above equation,

$$L^{(k_1 \ldots k_{n+1})}_{\sigma_{n+1}} = \sum_{\sigma_n \lambda_1} (-1)^{1+\sigma_n} [k_{n+1}] \begin{pmatrix} k_n & 1 & k_{n+1} \\ \sigma_n & -\lambda_1 & -\sigma_{n+1} \end{pmatrix} \frac{\partial A^{(k_1 \ldots k_n)}_{\sigma_n}}{\partial B^{(1)}_{\lambda_1}}. \tag{18b}$$

We note that the above results permit multiple differentiation of the tensor $\boldsymbol{A}$ with respect to variations of one or several tensorial fields. In particular, it results from (18*a*) that

$$\frac{\partial^2 A^{(k_1 \ldots k_n)}_{\sigma_n}}{\partial B^{(1)}_{\lambda_2} \partial B^{(1)}_{\lambda_1}} = \sum_{k_{n+1} k_{n+2}} (-1)^{\sigma_n + \sigma_{n+1}} [k_{n+1} k_{n+2}] \begin{pmatrix} k_n & 1 & k_{n+1} \\ \sigma_n & -\lambda_1 & -\sigma_{n+1} \end{pmatrix} \times \begin{pmatrix} k_{n+1} & 1 & k_{n+2} \\ \sigma_{n+1} & -\lambda_2 & -\sigma_{n+2} \end{pmatrix} L^{(k_1 \ldots k_{n+2})}_{\sigma_{n+2}}. \tag{19}$$

4. Differentiation of a tensor, arising from the coupling of *n* identical vectors, with respect to one of the vectors

In the preceding section, we discussed the case of differentiation of a tensor of rank *n* with respect to a tensor of rank *m*, when the result of the operation of differentiation had to be written in a basis of the order $n+m$. We shall now consider a tensor of the rank n constructed from components of a vector $A^{(1)}$ in conformity with the rule of coupling (6). The latter leads to

$$A^{(k_1\ldots k_n)}_{\sigma_n} = \sum_{\substack{\alpha_2\ldots\alpha_n \\ \sigma_1\ldots\sigma_{n-1}}} \prod_{i=1}^{n-1} (-1)^{k_i-1+\sigma_{i+1}}[k_{i+1}] \begin{pmatrix} k_i & 1 & k_{i+1} \\ \sigma_i & \alpha_{i+1} & -\sigma_{i+1} \end{pmatrix} A^{(1)}_{\alpha_i} A^{(1)}_{\alpha_n}. \tag{20}$$

Let us have a look at the derivative of the tensor (20) taken with respect to an arbitrary component of $A^{(1)}$. We obtain

$$\frac{\partial A^{(k_1\ldots k_n)}_{\sigma_n}}{\partial A^{(1)}_\lambda} = \sum_{\substack{\alpha_2\ldots\alpha_n \\ \sigma_1\ldots\sigma_{n-1}}} N \left(\delta_{\lambda\alpha_n} + A^{(1)}_{\alpha_n} \frac{\partial}{\partial A^{(1)}_\lambda} \right) \prod_{i=1}^{n-1} A^{(1)}_{\alpha_i} \tag{21}$$

where we have introduced the following notation:

$$N = \prod_{i=1}^{n-1} (-1)^{k_i-1+\sigma_{i+1}}[k_{i+1}] \begin{pmatrix} k_i & 1 & k_{i+1} \\ \sigma_i & \alpha_{i+1} & -\sigma_{i+1} \end{pmatrix}.$$

In deriving (21), we have made use of the relation

$$\partial A^{(1)}_{\alpha_n} / \partial A^{(1)}_\lambda = \delta_{\alpha_n\lambda}. \tag{22}$$

The latter results immediately from (18a) since differentiation of a vector with respect to itself leads to an irreducible spherical tensor of rank 0.

The operation of differentiation in the right-hand term of (21) can be rewritten as follows by having recourse to (22):

$$\frac{\partial}{\partial A^{(1)}_\lambda} \prod_{i=1}^{n-1} A^{(1)}_{\alpha_i} = \sum_{i=1}^{n-1} \delta_{\alpha_i\lambda} \prod_{j=1}^{i-1} A^{(1)}_{\alpha_j} \prod_{k=i+1}^{n-1} A^{(1)}_{\alpha_k} \tag{23}$$

whence, by (6),

$$\begin{aligned} \frac{\partial}{\partial A^{(1)}_\lambda} \prod_{i=1}^{n-1} A^{(1)}_{\alpha_i} = \sum_{i=1}^{n-1} \delta_{\alpha_i\lambda} \sum_{l_1\ldots l_{n-2}} \prod_{j=1}^{i-2} (-1)^{l_j-1+\lambda_{j+1}}[l_{j+1}] \\ \times \begin{pmatrix} l_j & 1 & l_{j+1} \\ \lambda_j & \alpha_{j+1} & -\lambda_{j+1} \end{pmatrix} \prod_{k=i+1}^{n-1} (-1)^{l_{k-2}-1+\lambda_{k-1}}[l_{k-1}] \\ \times \begin{pmatrix} l_{k-2} & 1 & l_{k-1} \\ \lambda_{k-2} & \alpha_k & -\lambda_{k-1} \end{pmatrix} A^{(l_1\ldots l_{n-2})}_{\lambda_{n-2}}. \end{aligned} \tag{24}$$

On insertion of (24) into (21) and on carrying out the necessary summations in accordance with formula (16), we finally obtain

$$\frac{\partial A^{(k_1\ldots k_n)}_{\sigma_n}}{\partial A^{(1)}_\lambda} = (-1)^{k_n+\sigma_n}[k_n] \sum_{l_1\ldots l_{n-1}} \begin{pmatrix} 1 & l_{n-1} & k_n \\ \lambda & \lambda_{n-1} & -\sigma_n \end{pmatrix} \Gamma(k_1 \ldots k_n l_1 \ldots l_{n-1}) A^{(l_1\ldots l_{n-1})}_{\lambda_{n-1}} \tag{25}$$

where

$$\begin{aligned} \Gamma(k_1 \ldots k_n l_1 \ldots l_{n-1}) \\ = (-1)^{k_n} \prod_{i=1}^{n-2} (-1)^{k_{i+2}+l_{i+1}}[k_{i+1}l_{i+1}] \begin{Bmatrix} 1 & l_{i+1} & k_{i+2} \\ 1 & k_{i+1} & l_i \end{Bmatrix} \\ + \sum_{i=1}^{n-1} \prod_{j=i}^{n-2} (-1)^{k_{j+1}+l_{j+1}}[k_{j+1}l_{j+1}] \begin{Bmatrix} 1 & l_{j+1} & k_{j+2} \\ 1 & k_{j+1} & l_j \end{Bmatrix} \prod_{m=1}^{i} \delta_{k_m l_m}. \end{aligned} \tag{26}$$

For $n=1$ we have $\Gamma(k_1 l_0)=1$, and differentiation of (25) gives a result which is in agreement with (22). We see from (25) that the expression of the result of a differentiation like this does not involve a basis of an order higher than the basis employed when constructing the tensor $A^{(k_1 \dots k_n)}$. Differentiation of the type (25) will lead to a reduction of the order of the basis.

From (25), it results that

$$\frac{\partial A^{(1k_2)}_{\sigma_2}}{\partial A^{(1)}_{\lambda}}=(-1)^{\sigma_2}[k_2](1+(-1)^{k_2})\begin{pmatrix}1 & 1 & k_2\\ \lambda & \sigma_1 & -\sigma_2\end{pmatrix}A^{(1)}_{\sigma_1}. \tag{27}$$

The preceding result (27) enables us to draw the significant conclusion that differentiation of an antisymmetric tensor of the second rank gives zero. Obviously, twofold differentiation of such a tensor gives the same. From (25), we obtain for $n=3$ that

$$\frac{\partial A^{(1k_2k_3)}_{\sigma_3}}{\partial A^{(1)}_{\lambda}}=\sum_{l_2}(-1)^{l_2+k_3+\sigma_3}(1+(-1)^{k_2})[k_2k_3l_2]\begin{pmatrix}1 & l_2 & k_3\\ \lambda & \sigma_2 & -\sigma_3\end{pmatrix}$$
$$\times\begin{Bmatrix}1 & k_3 & l_2\\ 1 & 1 & k_2\end{Bmatrix}A^{(1l_2)}_{\sigma_2}+(-1)^{k_3+\sigma_3}[k_3]\begin{pmatrix}1 & k_2 & k_3\\ \lambda & \sigma_2 & -\sigma_3\end{pmatrix}A^{(1k_2)}_{\sigma_2}. \tag{28}$$

If the tensor of rank 3 is antisymmetric in two indices (i.e. $k_2=1$), differentiation of the type (28) gives

$$\frac{\partial A^{(11k_3)}_{\sigma_3}}{\partial A^{(1)}_{\lambda}}=(-1)^{k_3+\sigma_3}[k_3]\begin{pmatrix}1 & 1 & k_3\\ \lambda & \sigma_2 & -\sigma_3\end{pmatrix}A^{(11)}_{\sigma_2} \tag{29}$$

and we obtain an antisymmetric tensor corresponding to a cartesian tensor of rank 2.

The preceding discussion shows that differentiation of antisymmetric tensors as well as differentiation with respect to antisymmetric tensors has to be performed very cautiously. Thus, for example, the result of differentiation of an arbitrary tensor with respect to an antisymmetric tensor of the second rank is indeterminate.

5. Applications

5.1. The polarisability tensor of rank 2 of an isolated molecule

We shall now apply the differential formulae derived above to the definition of some well known currently used tensorial quantities. Let us consider (18*b*) for $n=1$, assuming the tensor $\boldsymbol{A}$ to stand for the dipole moment $\boldsymbol{m}$ induced in a molecule and $\boldsymbol{B}$ to represent the vector of the electric field $\boldsymbol{E}$ interacting with the molecule. Accordingly

$$L^{(k_2)}_{\sigma_2}=\sum_{\sigma_1\lambda_1}(-1)^{1+\sigma_1}[k_2]\begin{pmatrix}1 & 1 & k_2\\ \sigma_1 & -\lambda_1 & -\sigma_2\end{pmatrix}\frac{\partial m^{(1)}_{\sigma_1}}{\partial E^{(1)}_{\lambda_1}}. \tag{30a}$$

We write the components of the spherical vector of the dipole moment as follows (Kaźmierczak 1985):

$$\boldsymbol{m}^{(1)}=\frac{1}{\sqrt{3}}\sum_{l}(-1)^{l}[l](\boldsymbol{a}^{(l)}\otimes\boldsymbol{E}^{(1)})^{(1)} \tag{31}$$

with $\boldsymbol{a}^{(l)}$ the spherical tensor of linear (second-rank) molecular polarisability. On inserting (31) into (30*a*) and applying (25) with $n=1$, or (22), we obtain

$$\boldsymbol{L}^{(k_2)}=\boldsymbol{a}^{(l)}\delta_{k_2 l}. \tag{30b}$$

Thus, the polarisability tensor $\boldsymbol{a}$ is defined as follows in spherical coordinates:

$$a^{(k_2)}_{\sigma_2} = \sum_{\sigma_1\lambda_1} (-1)^{1+\sigma_1}[k_2]\begin{pmatrix} 1 & 1 & k_2 \\ \sigma_1 & -\lambda_1 & -\sigma_2 \end{pmatrix}\frac{\partial m^{(1)}_{\sigma_1}}{\partial E^{(1)}_{\lambda_1}} \tag{32}$$

corresponding to $a_{ij} = \partial m_i/\partial E_j$ in cartesian coordinates (Kielich 1981).

5.2. *The tensor of interaction-induced polarisability of order 2 in the* DID *approximation*

Equation (32) allows us to determine the spherical tensor of interaction-induced polarisability of order 2 if the dipole moment $\boldsymbol{m}$ is induced as the result of interactions between the molecules. In accordance with (32), we write the tensor in question, for a molecular sample interacting as a whole with the external field of strength $\boldsymbol{E}$, as follows:

$$\Delta\Pi^{(k_2)}_{\sigma_2} = \sum_{i}^{N}\sum_{\sigma_1\lambda_1} (-1)^{1+\sigma_1}[k_2]\begin{pmatrix} 1 & 1 & k_2 \\ \sigma_1 & -\lambda_1 & -\sigma_2 \end{pmatrix}\frac{\partial\, {}^iM^{(1)}_{\sigma_1}}{\partial E^{(1)}_{\lambda_1}} \tag{33}$$

where summation over 'i' extends over all N molecules of the sample and the induced dipole moment is expressed by

$${}^i\boldsymbol{M}^{(1)} = \frac{1}{\sqrt{3}}\sum_{l_1} (-1)^{l_1}[l_1]({}^i\boldsymbol{a}^{(l_1)} \otimes {}^i\boldsymbol{F}^{(1)})^{(1)}. \tag{34}$$

Above, ${}^i\boldsymbol{F}$ is the vector of the molecular electric field produced at the centre of the ith molecule by the dipole moments induced in the other $(N-1)$ molecules of the sample (Kielich 1981):

$${}^i\boldsymbol{F}^{(1)} = -\left(\frac{5}{3}\right)^{1/2}\sum_{j\neq i}^{N}(\boldsymbol{T}^{(2)}(\boldsymbol{r}_{ij}) \otimes {}^j\boldsymbol{m}^{(1)})^{(1)} \tag{35}$$

where $\boldsymbol{T}(\boldsymbol{r}_{ij})$ is the tensor of dipole-dipole interaction dependent on the distance r_{ij} separating the molecules. In the lowest order of approximation of dipole-induced dipole (DID) interaction, the dipole moment ${}^j\boldsymbol{m}^{(1)}$ is given by (31).

Now, considering the form of the dipole moment (34), we have by (33) (see appendix):

$$\Delta\boldsymbol{\Pi}^{(k_2)} = \sum_{i}^{N}\sum_{k_1 l_1} (-1)^{k_2}[k_1 l_1]\begin{Bmatrix} k_1 & l_1 & k_2 \\ 1 & 1 & 1 \end{Bmatrix}({}^i\boldsymbol{a}^{(l_1)} \otimes {}^i\boldsymbol{L}^{(k_1)})^{(k_2)} \tag{36a}$$

where ${}^i\boldsymbol{L}$ is the tensor we get by differentiation of the molecular field vector ${}^i\boldsymbol{F}$ with respect to variations of the electric field $\boldsymbol{E}$. With regard to (35) as well as (A7) and (30b), we obtain (Kaźmierczak 1982):

$${}^i\boldsymbol{L}^{(k_1)} = -\sqrt{5}\sum_{j\neq i}^{N}\sum_{l_2} (-1)^{k_1}[l_2]\begin{Bmatrix} l_2 & 2 & k_1 \\ 1 & 1 & 1 \end{Bmatrix}(\boldsymbol{T}^{(2)}(\boldsymbol{r}_{ij}) \otimes {}^j\boldsymbol{a}^{(l_2)})^{(k_1)}. \tag{37}$$

Finally, on insertion of (37) into (36a), the interaction-induced polarisability tensor of second rank of the molecular sample in the lowest order of the DID approximation takes the following form (Kaźmierczak and Bancewicz 1984, De Santis and Sampoli 1984):

$$\Delta\boldsymbol{\Pi}^{(k_2)} = -\sqrt{5}\sum_{\substack{i,j \\ (i\neq j)}}^{N}\sum_{l_1 l_2 k_1} (-1)^{k_1+k_2}[k_1 l_1 l_2]\begin{Bmatrix} l_1 & k_2 & k_1 \\ 1 & 1 & 1 \end{Bmatrix}$$
$$\times\begin{Bmatrix} 2 & l_2 & k_1 \\ 1 & 1 & 1 \end{Bmatrix}({}^i\boldsymbol{a}^{(l_1)} \otimes (\boldsymbol{T}^{(2)}(\boldsymbol{r}_{ij}) \otimes {}^j\boldsymbol{a}^{(l_2)})^{(k_1)})^{(k_2)}. \tag{36b}$$

The result (36*b*), in cartesian coordinates, corresponds to the earlier formula (Kielich and Woźniak 1974, Kielich 1981):

$$\Delta\boldsymbol{\Pi} = -\sum_{\substack{i,j \\ (i\neq j)}}^{N} {}^{i}\boldsymbol{a}\cdot\boldsymbol{T}(\boldsymbol{r}_{ij})\cdot{}^{j}\boldsymbol{a}.$$

Hitherto, in papers dealing with the tensor $\Delta\boldsymbol{\Pi}$, its spherical representation has been derived by direct transformation of the preceding formula to a system of spherical coordinates.

5.3. *The third rank tensor of polarisability (hyperpolarisability) of an isolated molecule*

The spherical tensor of the electric dipole moment, induced in an isolated molecule in the process of two-photon polarisation, can be expressed by the equation (Ożgo 1975):

$$\boldsymbol{M}^{(1)} = \frac{1}{2\sqrt{3}}\sum_{lb}[l](\boldsymbol{B}^{(b,l)}\otimes\boldsymbol{E}^{(b)})^{(1)} \tag{38}$$

to which there corresponds the following expression in cartesian coordinates (Kielich 1980):

$$\boldsymbol{M} = \tfrac{1}{2}\boldsymbol{B}:\boldsymbol{EE}.$$

In (38) $\boldsymbol{B}^{(b,l)}$ is the non-symmetrised spherical tensor of molecular hyperpolarisability, and $\boldsymbol{E}^{(b)}$ corresponds to the dyad $\boldsymbol{EE}$ composed of the vector of the external electric field with which the molecule is at interaction. Assuming the tensor to be symmetric ($b\neq 1$), we have by (38)

$$\frac{\partial^2 M_\alpha^{(1)}}{\partial E_{\lambda_2}^{(1)}\partial E_{\lambda_1}^{(1)}} = \sum_{lb\lambda\beta}(-1)^{l-b+\alpha+\beta}[bl]\begin{pmatrix} l & b & 1 \\ \lambda & \beta & -\alpha\end{pmatrix}\begin{pmatrix} 1 & 1 & b \\ \lambda_1 & \lambda_2 & -\beta\end{pmatrix} B_\lambda^{(b,l)} \tag{39a}$$

where we have made use of the relations (22) and (27). At the same time, from (19), we find that

$$\frac{\partial^2 M_\alpha^{(1)}}{\partial E_{\lambda_2}^{(1)}\partial E_{\lambda_1}^{(1)}} = \sum_{k_2k_3}(-1)^{\lambda_1}[k_2k_3]\begin{pmatrix} 1 & 1 & k_2 \\ \alpha & -\lambda_1 & -\sigma_2\end{pmatrix}\begin{pmatrix} k_2 & 1 & k_3 \\ \sigma_2 & -\lambda_2 & -\sigma_3\end{pmatrix} L_{\sigma_3}^{(k_2k_3)}. \tag{39b}$$

On combining (39*a*) and (39*b*) and on summation over the 3*j* coefficients, applying (16) and the orthogonality relation for these coefficients, we obtain

$$\boldsymbol{B}^{(bk_3)} = \sum_{k_2}(-1)^{k_2+b+1}[k_2b]\begin{Bmatrix} 1 & k_2 & k_3 \\ 1 & b & 1\end{Bmatrix}\boldsymbol{L}^{(k_2k_3)} \tag{40a}$$

whence we arrive directly at the definition of the hyperpolarisability tensor $\boldsymbol{B}^{(b,k_3)}$ by way of the second derivative of the dipole moment (38) on replacing $\boldsymbol{L}^{(k_2k_3)}$ by the expression obtained on inversion of (39*b*):

$$B_{\sigma_3}^{(bk_3)} = \sum_{\lambda_1\lambda_2\alpha\beta}(-1)^{k_3-b+\sigma_3}[bk_3]\begin{pmatrix} 1 & b & k_3 \\ \alpha & -\beta & -\sigma_3\end{pmatrix}\begin{pmatrix} 1 & 1 & b \\ \lambda_1 & \lambda_2 & -\beta\end{pmatrix}\frac{\partial^2 M_\alpha^{(1)}}{\partial E_{\lambda_2}^{(1)}\partial E_{\lambda_1}^{(1)}}. \tag{40b}$$

It is easy to check that the preceding result is identical with that obtained by inversion of (39*a*).

5.4. The dipole-quadrupole polarisability of a molecule

The preceding examples illustrate different applications of the formula (17) for $m=1$. Here, we shall deal with the problem of differentiation with respect to a tensor of rank 2 ($m=2$). To this aim, we consider the tensor of dipole-quadrupole molecular polarisability $A_{i,jk}$ used for the description of the mechanism of induction of a molecular dipole moment by the gradient of an external electric field, as well as the induction of a quadrupole moment (Tabisz 1979):

$$m_i = \tfrac{1}{3} A_{i,jk} \nabla_j E_k \tag{41}$$

$$Q_{ij} = A_{ij,k} E_k. \tag{42}$$

To establish the relationship between $\boldsymbol{A}$ and $\partial \boldsymbol{m}/\partial(\nabla \boldsymbol{E})$, it suffices to put $n=1$ and $m=2$ in (17). We immediately obtain

$$\frac{\partial m_{\sigma_1}^{(1)}}{\partial(\nabla E)_{\lambda_2}^{(1l_2)}} = \tfrac{1}{3} \sum_{k_2 k_3} (-1)^{k_3+\sigma_1} [l_2 k_2 k_3] \begin{pmatrix} 1 & l_2 & k_3 \\ \sigma_1 & -\lambda_2 & -\sigma_3 \end{pmatrix} \begin{Bmatrix} 1 & l_2 & k_3 \\ 1 & k_2 & 1 \end{Bmatrix} A_{\sigma_3}^{(1k_2k_3)}. \tag{43}$$

Equation (43) gives the tensor $\boldsymbol{A}^{(1k_2k_3)}$, albeit in non-symmetrised form. It is composed in accordance with the rule (7), i.e. according to the scheme

$$\boldsymbol{A}^{(1k_2k_3)} = [(\boldsymbol{A}^{(1)} \otimes \boldsymbol{A}^{(1)})^{(k_2)} \otimes \boldsymbol{A}^{(1)}]^{(k_3)}.$$

Thus, the above scheme corresponds to the construction of the tensor $A_{ij,k}$ of (42) and its relation to $\partial \boldsymbol{Q}/\partial \boldsymbol{E}$ can be obtained directly from (18*a*). On the other hand, from the form in which (41) is expressed, it results that the tensor $A_{i,jk}$ has to be coupled according to the scheme

$$\boldsymbol{A}^{(1k_2'k_3)} = [\boldsymbol{A}^{(1)} \otimes (\boldsymbol{A}^{(1)} \otimes \boldsymbol{A}^{(1)})^{(k_2')}]^{(k_3)}.$$

Thus, we come upon the problem of symmetrisation of the tensors obtained by way of differentiation. In the present example, we no more than state the problem.

The relation between the preceding coupling schemes is the following (Biedenharn and Louck 1981, Varshalovich *et al* 1975):

$$\boldsymbol{A}^{(1k_2k_3)} = (-1)^{1+k_3} \sum_{k_2'} [k_2 k_2'] \begin{Bmatrix} 1 & 1 & k_2 \\ 1 & k_3 & k_2' \end{Bmatrix} \boldsymbol{A}^{(1k_2'k_3)}. \tag{44}$$

On insertion of (44) into (43) and on summation over k_2 we obtain

$$\frac{\partial m_{\sigma_1}^{(1)}}{\partial(\nabla E)_{\lambda_2}^{(1l_2)}} = \tfrac{1}{3} \sum_{k_3} (-1)^{1+\sigma_1} [k_3] \begin{pmatrix} 1 & l_2 & k_3 \\ \sigma_1 & -\lambda_2 & -\sigma_3 \end{pmatrix} A_{\sigma_3}^{(1l_2k_3)} \tag{45}$$

whence, on inversion of the equation, we obtain

$$A_{\sigma_3}^{(1l_2k_3)} = 3 \sum_{\sigma_1 \lambda_2} (-1)^{1+\sigma_1} [k_3] \begin{pmatrix} 1 & l_2 & k_3 \\ \sigma_1 & -\lambda_2 & -\sigma_3 \end{pmatrix} \frac{\partial m_{\sigma_1}^{(1)}}{\partial(\nabla E)_{\lambda_2}^{(1l_2)}}. \tag{46}$$

The spherical tensor $\boldsymbol{A}^{(1l_2k_3)}$ thus obtained now possesses the required symmetry in accordance with the symmetry of the cartesian tensor $A_{i,jk}$.

Acknowledgment

The author wishes to express his indebtedness to Professor S Kielich for numerous highly valuable discussions and suggestions throughout the present investigation.

Appendix

The following tensorial product

$$\boldsymbol{A}^{(1)}=(\boldsymbol{C}^{(l)}\otimes\boldsymbol{D}^{(1)})^{(1)} \tag{A1}$$

is to be differentiated with respect to a tensor $\boldsymbol{B}^{(1)}$. In (A1) let $\boldsymbol{D}^{(1)}$ alone be a function of $\boldsymbol{B}^{(1)}$. With regard to (5*a*), we obtain

$$\frac{\partial A_\alpha^{(1)}}{\partial B_\beta^{(1)}}=\sqrt{3}\sum_{\gamma\delta}(-1)^{l-1+\alpha}\begin{pmatrix} l & 1 & 1\\ \gamma & \delta & -\alpha\end{pmatrix} C_\gamma^{(l)}\frac{\partial D_\delta^{(1)}}{\partial B_\beta^{(1)}}. \tag{A2}$$

On applying (18*a*) with $n=1$ to the derivative in the right-hand term of (A2), we obtain

$$\frac{\partial A_\alpha^{(1)}}{\partial B_\beta^{(1)}}=\sqrt{3}\sum_{k\gamma\delta}(-1)^{l+\gamma}[k]\begin{pmatrix} l & 1 & 1\\ \gamma & \delta & -\alpha\end{pmatrix}\begin{pmatrix} 1 & 1 & k\\ \delta & -\beta & -\sigma\end{pmatrix} C_\gamma^{(l)}L_\sigma^{(k)}. \tag{A3}$$

On applying the following formula (Varshalovich *et al* 1975):

$$\sum_{\varepsilon}(-1)^{e-\varepsilon}\begin{pmatrix} a & b & e\\ \alpha & \beta & -\varepsilon\end{pmatrix}\begin{pmatrix} e & d & c\\ \varepsilon & \delta & \gamma\end{pmatrix}$$

$$=\sum_{n\nu}(-1)^{n-\nu}[n]^2\begin{pmatrix} a & c & n\\ \alpha & \gamma & -\nu\end{pmatrix}\begin{pmatrix} n & d & b\\ \nu & \delta & \beta\end{pmatrix}\begin{Bmatrix} b & d & n\\ c & a & e\end{Bmatrix} \tag{A4}$$

we reduce (A3) to the form

$$\frac{\partial A_\alpha^{(1)}}{\partial B_\beta^{(1)}}=\sqrt{3}\sum_{kh}(-1)^{l+h+1+\alpha}[hk]\begin{pmatrix} 1 & 1 & h\\ \alpha & -\beta & -\chi\end{pmatrix}\begin{Bmatrix} k & l & h\\ 1 & 1 & 1\end{Bmatrix}(C^{(l)}\otimes L^{(k)})_\chi^{(h)}. \tag{A5}$$

On multiplying both terms of (A5) by the coefficient

$$\begin{pmatrix} 1 & 1 & r\\ \alpha & -\beta & -\rho\end{pmatrix}$$

and on summation over α and β, (A5) becomes

$$[h]\sum_{\alpha\beta}(-1)^{1+\alpha}\begin{pmatrix} 1 & 1 & h\\ \alpha & -\beta & -\chi\end{pmatrix}\frac{\partial A_\alpha^{(1)}}{\partial B_\beta^{(1)}}=\sqrt{3}\sum_{k}(-1)^{l+h}[k]\begin{Bmatrix} k & l & h\\ 1 & 1 & 1\end{Bmatrix}(\boldsymbol{C}^{(l)}\otimes\boldsymbol{L}^{(k)})_\chi^{(h)} \tag{A6}$$

whence, taking into account that the left-hand term of (A6) is equal to $L_\chi^{(h)'}$ (cf equation (18*b*)), we finally obtain

$$\boldsymbol{L}^{(h)'}=\sqrt{3}\sum_{k}(-1)^{l+h}[k]\begin{Bmatrix} k & l & h\\ 1 & 1 & 1\end{Bmatrix}(\boldsymbol{C}^{(l)}\otimes\boldsymbol{L}^{(k)})^{(h)}. \tag{A7}$$

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