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Spherical Harmonic Expansion of the Fokker-Planck Equation

By

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Summary: The Fokker-Planck equation is expanded in terms of spherical harmonics. The result is quite general without any restriction on the distortion of the velocity distribution function. A corresponding expression is also presented for the case where electric and magnetic fields exist.

§ 1 INTRODUCTION

Over the extended fields of plasma physics, Coulomb interaction plays an important role in determining the macroscopic properties of the system. The Fokker-Planck equation is well known for a wide range of applicability and a substantial portion is usually devoted to its derivation in current standard textbooks on plasma physics. The equation takes the form of integro-differential (second order) equation in three dimensional velocity space. Direct utilization of the form, however, is not generally pertinent to numerical calculation even with use of the developed computational facilities of these days. (The literatures on the computational works to date are seen in the recent review article by Killeen *et al.* [*1*].)

In practical applications, a variety of approximations have been made according to the purposes. Most common simplification is made by assuming for a deviation of the distribution function from the equilibrium being small and accordingly by linearizing the equation. If we consider the problem in which no simplification due to symmetry consideration is possible and in which there is no restriction for the applied field strengths, an expansion of the equation may be devised in terms of some systems of orthogonal functions of appropriate variables in velocity space.

Cartesian tensor expansion was successful only in the case of the linearized version where the terms containing the zeroth order distribution function alone were retained [*2*]. The failure of further developement was due to the general difficulty of angular integration in his tensor formalism. It is the purpose of this paper to apply a spherical harmonic expansion for an arbitrary velocity distribution function and finally to derive an expression involving only one scalar variable, *i.e.*, the magnitude of the velocity vector. Although the spherical harmonic expansion is in a rigorous sense equivalent to the Cartesian tensor formalism [*3*], the former has more advantage than the latter, since the spherical harmonics have been extensively studied in the past from the necessity in atomic and nuclear sciences.

To make our standpoint clear, we consider the Boltzmann equation assuming a uniformity in space,

$$\frac{\partial f_i}{\partial t}+\frac{z_i e}{m_i}(\boldsymbol{E}+\boldsymbol{v}\times\boldsymbol{B})\cdot\nabla_v f_i=\left(\frac{\delta f_i}{\delta t}\right)_{F-P}+(\cdots)\,, \tag{1}$$

where $\boldsymbol{v}$ is the velocity vector, f_i the velocity distribution function of the i-th kind of particles normalized such that $\int f_i(\boldsymbol{v})d\boldsymbol{v}$ gives the number density in a unit volume of space, and $z_i e$ and m_i are the charge and mass, respectively. $\boldsymbol{E}$ and $\boldsymbol{B}$ are the electric and magnetic fields, respectively. $\left(\frac{\delta f_i}{\delta t}\right)_{F-P} d\boldsymbol{v}$ gives a net gain of the i-th particles into the phase space $\boldsymbol{v}$ to $\boldsymbol{v}+d\boldsymbol{v}$ due to the Coulomb scattering in a unit time. (Generally this is a simple summation of the corresponding Fokker-Planck terms for the respective pairs of i- and j-th particles, including like-particle scattering.) $(\cdots)$ implies all other collisional processes allowing the production and loss of the i-th particles. In this category a number of collisional processes involving neutral and ionized species are included: elastic scattering, rotational and vibrational excitation, electronic excitation, and the corresponding de-excitations, ionization, secondary electron ejection, recombination, chemical reaction, photoexcitation, photodissociation, photoionization, and so forth. Each elementary process as mentioned requires a quite unique treatment in the respective formulation. Although a formal description is not always difficult, a rigorous derivation of an expression with one variable alone seems to be desparately complicated for certain types of processes. The reason is mainly due to the finite energy losses (which are not always small) and due to the complicated scattering laws resulting from the diverse natures of interaction potentials. From such a point of view, it might as well be said that the Fokker-Planck equation is more accessible since the scattering law to be considered in this case is quite simple, although the derivation of the final expression still requires considerable manipulations as is shown in the subsequent section.

§ 2 Formulation

We consider two types of particles specified by *1* and *2*. Let the mass, charge and velocity distribution function for particle *1* be m_1, $z_1 e$ and $f(\boldsymbol{v})$, respectively and for particle *2*, m_2, $z_2 e$ and $F(\boldsymbol{v})$, respectively. Then the Fokker-Planck equation can be written as follows [4],

$$\left(\frac{\delta f}{\delta t}\right)_{F-P}=\Gamma\left[4\pi\frac{m_1}{m_2}F(\boldsymbol{v})f(\boldsymbol{v})+\frac{m_2-m_1}{m_2+m_1}\nabla\mathscr{H}\cdot\nabla f+\frac{\nabla\nabla\mathscr{G}:\nabla\nabla f}{2}\right], \tag{2}$$

where

$$\Gamma=4\pi\left(\frac{z_1 z_2 e^2}{m_e}\right)^2 ln\,\Lambda.$$

$-e$ and m_e are the charge and mass of electron and Λ is the quantity customarily called 'Coulomb Λ'. The symbol : shows a tensor contraction. $\mathscr{H}$ and $\mathscr{G}$ are the Rosenbluth potentials [5] defined as

$$\mathscr{H}(\boldsymbol{v})=\frac{m_1+m_2}{m_2}\int\frac{F(V)}{\mathscr{V}}dV, \tag{3}$$

and

$$\mathscr{G}(\boldsymbol{v})=\int F(V)\mathscr{V}dV, \tag{4}$$

where $\mathscr{V}=|\boldsymbol{v}-\boldsymbol{V}|$.

We use the spherical coordinate system whose polar axis is arbitrarily fixed in space and expand $f(\boldsymbol{v})$ and $F(\boldsymbol{v})$ in terms of spherical harmonics* as follows,

$$f(\boldsymbol{v})=\sum_{lm}f_{lm}(v)Y_{lm}(\theta,\phi) \tag{5}$$

$$F(\boldsymbol{v})=\sum_{LM}F_{LM}(v)Y_{LM}(\theta,\phi), \tag{6}$$

where θ and ϕ are the polar and azimuthal angles, respectively. The last term in eq. (2) can be expressed in tensor notation as

$$\nabla\nabla\mathscr{G}:\nabla\nabla f=\mathscr{G}_{,\mu\nu}g^{\mu\omega}g^{\nu\tau}f_{,\omega\tau} \tag{7}$$

where the symbol , indicates a covariant derivative defined by

$$f_{,\omega\tau}=\frac{\partial^2 f}{\partial u^\omega\partial u^\tau}-\left\{{\sigma \atop \omega\tau}\right\}\frac{\partial f}{\partial u^\sigma}. \tag{8}$$

Here, (u^1, u^2, u^3) represents a general coordinate system and $(\mu, \nu, \sigma, \tau, \omega)$ the indices. $g^{\mu\omega}$ and $\left\{{\sigma \atop \omega\tau}\right\}$ are the metric tensor and the Christoffel symbol, respectively often used in the tensor analysis.

Specialization of $g^{\mu\omega}$ and $\left\{{\sigma \atop \omega\tau}\right\}$ to our present coordinate system and their substitution into eq. (7) lead to

$$\begin{aligned}\nabla\nabla\mathscr{G}:\nabla\nabla f=&\frac{\partial^2\mathscr{G}}{\partial v^2}\frac{\partial^2 f}{\partial v^2}+\frac{1}{v^4\sin^4\theta}\left(\frac{\partial^2\mathscr{G}}{\partial\phi^2}+v\sin^2\theta\frac{\partial\mathscr{G}}{\partial v}+\sin\theta\cos\theta\frac{\partial\mathscr{G}}{\partial\theta}\right)\\&\times\left(\frac{\partial^2 f}{\partial\phi^2}+v\sin^2\theta\frac{\partial f}{\partial v}+\sin\theta\cos\theta\frac{\partial f}{\partial\theta}\right)\\&+\frac{2}{v^2}\left(\frac{\partial^2\mathscr{G}}{\partial v\partial\theta}-\frac{1}{v}\frac{\partial\mathscr{G}}{\partial\theta}\right)\left(\frac{\partial^2 f}{\partial v\partial\theta}-\frac{1}{v}\frac{\partial f}{\partial\theta}\right)\end{aligned} \tag{9}$$

* The definition of spherical harmonics, particularly in its phase factor, differs from book to book. Our definition obeys Rose [6]. $f(\boldsymbol{v})$ and $F(\boldsymbol{v})$ are real quantities, hence it follows the relations

$$f_{l,-m}=(-1)^m f^*_{lm} \quad\text{and}\quad F_{L,-M}=(-1)^M F^*_{LM}.$$

$$+\frac{2}{v^2\sin^2\theta}\left(\frac{\partial^2\mathcal{G}}{\partial v\partial\phi}-\frac{1}{v}\frac{\partial\mathcal{G}}{\partial\phi}\right)\left(\frac{\partial^2 f}{\partial v\partial\phi}-\frac{1}{v}\frac{\partial f}{\partial\phi}\right)$$
$$+\frac{2}{v^4\sin^2\theta}\left(\frac{\partial^2\mathcal{G}}{\partial\theta\partial\phi}-\cot\theta\frac{\partial\mathcal{G}}{\partial\phi}\right)\left(\frac{\partial^2 f}{\partial\theta\partial\phi}-\cot\theta\frac{\partial f}{\partial\phi}\right)$$
$$+\frac{1}{v^4}\left(\frac{\partial^2\mathcal{G}}{\partial\theta^2}+v\frac{\partial\mathcal{G}}{\partial v}\right)\left(\frac{\partial^2 f}{\partial\theta^2}+v\frac{\partial f}{\partial v}\right).$$

Next, the potentials $\mathcal{H}(\boldsymbol{v})$ and $\mathcal{G}(\boldsymbol{v})$ have to be expanded in terms of spherical harmonics. Since the procedures is based on a series of conventional techniques, we describe the outline only.

Firstly, relative velocity $\mathcal{V}=|\boldsymbol{v}-\boldsymbol{V}|$ and its reciprocal $\mathcal{V}^{-1}$ are expanded in terms of the Legendre function $P_N(\hat{\boldsymbol{v}}\cdot\hat{\boldsymbol{V}})$. With the help of the addition theorem for the spherical harmonics, the Legendre function is led to a form of

$$\frac{4\pi}{2N+1}\sum_n Y^*_{Nn}(\hat{\boldsymbol{V}})Y_{Nn}(\hat{\boldsymbol{v}}).$$

Substitution of the resulting expression and eq. (6) into eqs. (3) and (4) and subsequent integration over $\boldsymbol{V}$ lead to the following results.

Writing $\mathcal{H}(\boldsymbol{v})$ and $\mathcal{G}(\boldsymbol{v})$ as

$$\mathcal{H}(\boldsymbol{v})=\sum_{LM}H_{LM}(v)Y_{LM}(\hat{\boldsymbol{v}}) \tag{10}$$

and

$$\mathcal{G}(\boldsymbol{v})=\sum_{LM}G_{LM}(v)Y_{LM}(\hat{\boldsymbol{v}}), \tag{11}$$

one finds

$$H_{LM}(v)=\frac{m_1+m_2}{m_2}\frac{1}{2L+1}\frac{1}{v}(I^{LM}_L+J^{LM}_{-L-1}) \tag{12}$$

and

$$G_{LM}(v)=\frac{v}{(2L+1)(2L+3)}(I^{LM}_{L+2}+J^{LM}_{-L-1}) -\frac{v}{(2L-1)(2L+1)}(I^{LM}_L+J^{LM}_{-L+1}), \tag{13}$$

where

$$I^{LM}_\alpha=\frac{4\pi}{v^\alpha}\int_0^v F_{LM}(V)V^{\alpha+2}dV \tag{14}$$

and

$$J^{LM}_\alpha=\frac{4\pi}{v^\alpha}\int_v^\infty F_{LM}(V)V^{\alpha+2}dV. \tag{15}$$

The expanded form of eq. (11) allows one to write the term $\nabla\nabla\mathscr{G}:\nabla\nabla f$ in the following form;

$$
\begin{aligned}
&\nabla\nabla\mathscr{G}:\nabla\nabla f \\
&= \sum_{\substack{LM \\ lm}} \Bigg\{ \left(G''_{LM} f''_{lm} + \frac{2G'_{LM} f'_{lm}}{v^2} \right) |LM> \cdot |lm> \\
&\qquad\qquad + \frac{G_{LM} f_{lm}}{v^4} \cdot \frac{\partial^2}{\partial\theta^2} |LM> \cdot \frac{\partial^2}{\partial\theta^2} |lm> \\
&\quad + \frac{G'_{LM} f_{lm}}{v^3} |LM> \cdot \frac{\partial^2}{\partial\theta^2} |lm> + \frac{G_{LM} f'_{lm}}{v^3} \frac{\partial^2}{\partial\theta^2} |LM> \cdot |lm> \\
&\quad - \frac{G_{LM} f_{lm}}{v^4} (m^2 - 2mM) \frac{\cot\theta}{\sin^2\theta} \frac{\partial}{\partial\theta} |LM> \cdot |lm> \\
&\quad - \frac{G_{LM} f_{lm}}{v^4} (M^2 - 2Mm) \frac{\cot\theta}{\sin^2\theta} |LM> \cdot \frac{\partial}{\partial\theta} |lm> \\
&\quad - \left[\frac{G'_{LM} f_{lm}}{v^3} m^2 + \frac{G_{LM} f'_{lm}}{v^3} M^2 + 2mM \left(\frac{G'_{LM}}{v} - \frac{G_{LM}}{v^2} \right) \right. \\
&\qquad \left. \times \left(\frac{f'_{lm}}{v} - \frac{f_{lm}}{v^2} \right) \right] \frac{1}{\sin^2\theta} |LM> \cdot |lm> \\
&\quad + 2 \left(\frac{G'_{LM}}{v} - \frac{G_{LM}}{v^2} \right) \left(\frac{f'_{lm}}{v} - \frac{f_{lm}}{v^2} \right) \frac{\partial}{\partial\theta} |LM> \cdot \frac{\partial}{\partial\theta} |lm> \\
&\quad + \frac{G_{LM} f_{lm}}{v^4} \cot^2\theta \frac{\partial}{\partial\theta} |LM> \cdot \frac{\partial}{\partial\theta} |lm> \\
&\quad + \frac{G_{LM} f'_{lm}}{v^3} \cot\theta \frac{\partial}{\partial\theta} |LM> \cdot |lm> + \frac{G'_{LM} f_{lm}}{v^3} \cot\theta |LM> \cdot \frac{\partial}{\partial\theta} |lm> \\
&\quad + \frac{m^2 M^2}{v^4} G_{LM} f_{lm} \frac{1}{\sin^4\theta} |LM> \cdot |lm> - 2mM \frac{G_{LM} f_{lm}}{v^4} \frac{\cot^2\theta}{\sin^2\theta} |LM> \cdot |lm> \\
&\quad - 2mM \frac{G_{LM} f_{lm}}{v^4} \frac{1}{\sin^2\theta} \frac{\partial}{\partial\theta} |LM> \cdot \frac{\partial}{\partial\theta} |lm> \Bigg\}.^{**} \qquad (16)
\end{aligned}
$$

Similar expressions for the terms $\nabla\mathscr{H}\cdot\nabla f$ and $F(\boldsymbol{v})f(\boldsymbol{v})$ in eq. (2) can be derived more easily.

Next we write the Fokker-Planck equation (2) as

$$\left(\frac{\delta f}{\delta t} \right)_{F-P} = \sum_{pq} \left(\frac{\delta f}{\delta t} \right)_{pq} (v) Y_{pq}(\theta, \phi), \qquad (17)$$

i.e.,

$$\left(\frac{\delta f}{\delta t} \right)_{pq} = \int Y^*_{pq}(\theta, \phi) \left(\frac{\delta f}{\delta t} \right)_{F-P} d\Omega_{\theta,\phi}. \qquad (18)$$

** For the sake of brevity, $|lm>$ and superscript $'$ are used to indicate $Y_{lm}(\theta, \phi)$ and differentiation with respect to v, respectively.

Our final purpose is to derive the explicit expression for the coefficient (18). The subsequent calculation is quite tedious though the included manipulation is rather elementary. In Appendix the relations necessary for this purpose are listed. All the terms thus obtained, if appropriately arranged, eventually lead to a single integral $\int Y^{*}_{pq} Y_{LM} Y_{lm} d\Omega_{\theta,\phi}$ which is given in terms of the C-G (Clebsch-Gordan) coefficients. H_{LM} and G_{LM} and their first and second derivatives can be obtained according to the eqs. (12) and (13). Rearrangement of the resulting terms finally yields

$$\left(\frac{\delta f}{\delta t}\right)_{pq} = \Gamma \cdot \sum_{Ll} \sqrt{\frac{(2L+1)(2l+1)}{4\pi(2p+1)}}\, C(Llp|000) \times \sum_{Mm} C(Llp|Mmq)\left(\chi_1 \frac{\partial^2 f_{lm}}{\partial v^2} + \chi_2 \frac{\partial f_{lm}}{\partial v} + \chi_3 f_{lm}\right), \tag{19}$$

where

$$\chi_1 = \frac{1}{v}\left[\nu_{LL} I_L^{LM} - \nu_{L,L+2} I_{L+2}^{LM} + \nu_{L,-L-1} J_{-L-1}^{LM} - \nu_{L,-L+1} J_{-L+1}^{LM}\right] \tag{20}$$

$$\chi_2 = \frac{1}{v^2}\left[\left(-\frac{L+1}{2L+1}(1-\mu) + \varepsilon_{L,L}\right) I_L^{LM} - \varepsilon_{L,L+2} I_{L+2}^{LM} + \left(\frac{L}{2L+1}(1-\mu) + \varepsilon_{L,-L-1}\right) J_{-L-1}^{LM} - \varepsilon_{L,-L+1} J_{-L+1}^{LM}\right] \tag{21}$$

$$\chi_3 = 4\pi\mu F_{LM} + \frac{1}{v^3}\left[\left(-\frac{\lambda}{2(2L+1)}(1-\mu) + \eta_{L,L}\right) I_L^{LM} - \eta_{L,L+2} I_{L+2}^{LM} + \left(-\frac{\lambda}{2(2L+1)}(1-\mu) + \eta_{L,-L-1}\right) J_{-L-1}^{LM} - \eta_{L,-L+1} J_{-L+1}^{LM}\right]. \tag{22}$$

Here, ν, ε, and η are simple numerical constants given by

$$\nu_{L,\gamma} = -\frac{1}{2(2L+1)} \frac{\gamma(\gamma-1)}{2\gamma-1}, \tag{23}$$

$$\varepsilon_{L,\gamma} = -\frac{1}{2(2L+1)(2\gamma-1)}\{\lambda - L(L+1) + (\lambda-2)(\gamma-1)\} \tag{24}$$

and

$$\eta_{L,\gamma} = \frac{1}{2(2L+1)(2\gamma-1)}\left\{[\lambda - l(l+1)](\gamma-1) - \frac{\lambda(\lambda-2)}{4}\right\}, \tag{25}$$

where,

$$\lambda = p(p+1) - L(L+1) - l(l+1), \tag{26}$$

and μ is the mass ratio of both particles, i.e., $\mu = m_1/m_2$. As to the *C-G* coefficients appeared in eq. (19), one may refer to the textbook by Rose [6]. Take notice of the form of eq. (19) where M and m, projected angular momenta, do not appear explicitly except in the *C-G* coefficients[+)].

This can be understood from the Wigner-Eckart theorem that the matrix element of an irreducible tensor is separable into the two parts, geometric part and physical part.

We have thus completed the main purpose of this paper. Back to eq. (1), it may be useful to present the corresponding expression consistently obtainable from our spherical harmonic expansion for the case where the external electric and magnetic fields exist. For these fields we allow arbitrary directions. Indeed, one can adopt either of them as the polar axis of the coordinate system and obtain a slightly more simplified result. However, we do not take such a coordinate system considering the possibility that there might exist some other important symmetry directions in the actual problems characterized by macroscopic fluid direction, radiation insolation, design of experimental apparatus etc..

We define, $B^+ = B_x + iB_y$, $B^- = B_x - iB_y$, $E^+ = E_x + iE_y$ and $E^- = E_x - iE_y$, where i is the imaginary unit, and B_x, B_y, E_x and E_y show the x and y components in the Cartesian coordinates, respectively. (where the z axis is chosen along the polar axis in our present spherical harmonic expansion.) Then it follows that

$$
\begin{aligned}
\frac{\partial}{\partial t} f_{pq} + \frac{z_1 e}{m_1} \Big\{ & -iqB_z f_{pq} - \frac{i}{2} B^+ G_{pq} f_{p,q-1} - \frac{i}{2} B^- K_{pq} f_{p,q-1} \\
& + E_z \Big[B_{pq} \Big(f'_{p-1,q} - (p-1) \frac{f_{p-1,q}}{v} \Big) + A_{pq} \Big(f'_{p-1,q} + (p+2) \frac{f_{p+1,q}}{v} \Big) \Big] \\
& + E^+ \Big[\frac{D_{pq}}{2} \Big(f'_{p-1,q+1} - (p-1) \frac{f_{p-1,q+1}}{v} \Big) \\
& \qquad - \frac{C_{pq}}{2} \Big(f'_{p-1,q+1} + (p+2) \frac{f_{p+1,q+1}}{v} \Big) \Big] \\
& + E^- \Big[- \frac{F_{pq}}{2} \Big(f'_{p-1,q-1} - (p-1) \frac{f_{p-1,q-1}}{v} \Big) \\
& \qquad + \frac{E_{pq}}{2} \Big(f'_{p-1,q-1} + (p+2) \frac{f_{p+1,q-1}}{v} \Big) \Big] \Big\} \\
= \text{righthand side of eq. (19),} & \qquad (27)
\end{aligned}
$$

where the terms including f_{ij} should be retained only if the condition $i \geqq |j|$ is satisfied. The numerical constants A_{pq}, B_{pq}, C_{pq}, D_{pq}, E_{pq}, F_{pq}, G_{pq} and K_{pq} are explicitly given in Appendix.

[+] Thence, selection rules follow as $M + m = q$, $L + l + p =$ even integer, and the triangular relation $\Delta(Llp)$ (sum of any two of the quantities L, l, and p must not be less than the rest).

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APPENDIX

The relations necessary for showing that eq. (16) is expressible in terms of the products of two spherical harmonics alone are as follows;

$$\cos\theta\,|lm> = A_{lm}\,|l+1, m> + B_{lm}\,|l-1, m>, \tag{A·1}$$

$$m\cot\theta\,|lm> = -\frac{1}{2}G_{lm}e^{-i\phi}\,|l, m+1> - \frac{1}{2}K_{lm}e^{i\phi}\,|l, m-1>, \tag{A·2}$$

$$\frac{\partial}{\partial\theta}\,|lm> = \frac{1}{2}G_{lm}e^{-i\phi}\,|l, m+1> - \frac{1}{2}K_{lm}e^{i\phi}\,|l, m-1>, \tag{A·3}$$

$$e^{i\phi}\sin\theta\,|lm> = -C_{lm}\,|l+1, m+1> + D_{lm}\,|l-1, m+1>, \tag{A·4}$$

$$e^{-i\phi}\sin\theta\,|lm> = E_{lm}\,|l+1, m-1> - F_{lm}\,|l-1, m-1>. \tag{A·5}$$

Here, the numerical coefficients are defined by

$$A_{lm} = \sqrt{\frac{(l+m+1)(l-m+1)}{(2l+1)(2l+3)}}, \tag{A·6}$$

$$B_{lm} = \sqrt{\frac{(l+m)(l-m)}{(2l-1)(2l+1)}}, \tag{A·7}$$

$$C_{lm} = \sqrt{\frac{(l+m+1)(l+m+2)}{(2l+1)(2l+3)}}, \tag{A·8}$$

$$D_{lm} = \sqrt{\frac{(l-m)(l-m-1)}{(2l-1)(2l+1)}}, \tag{A·9}$$

$$E_{lm} = \sqrt{\frac{(l-m+1)(l-m+2)}{(2l+1)(2l+3)}}, \tag{A·10}$$

$$F_{lm} = \sqrt{\frac{(l+m)(l+m-1)}{(2l-1)(2l+1)}}, \tag{A·11}$$

$$G_{lm}=\sqrt{(l-m)(l+m+1)}\ , \tag{A·12}$$

$$K_{lm}=\sqrt{(l+m)(l-m+1)}\ . \tag{A·13}$$

The relations necessary for the derivation of eq. (27) are

$$\begin{aligned}\langle pq|\cos\theta\cos\phi\frac{\partial}{\partial\theta}-\frac{\sin\phi}{\sin\theta}\frac{\partial}{\partial\phi}|lm\rangle=\frac{1}{2}[(p-1)(F_{pq}-D_{pq})\\ +(p+2)(E_{pq}-C_{pq})],\end{aligned} \tag{A.14}$$

$$\begin{aligned}\langle pq|\cos\theta\sin\phi\frac{\partial}{\partial\theta}+\frac{\cos\phi}{\sin\theta}\frac{\partial}{\partial\phi}|lm\rangle=-\frac{i}{2}[(p-1)(F_{pq}+D_{pq})\\ +(p+2)(E_{pq}+C_{pq})],\end{aligned} \tag{A.15}$$

$$\langle pq|\sin\phi\frac{\partial}{\partial\theta}+\cos\phi\cot\theta\frac{\partial}{\partial\phi}|lm\rangle=-\frac{i}{2}[G_{pq}+K_{pq}], \tag{A·16}$$

and

$$\langle pq|\cos\phi\frac{\partial}{\partial\theta}-\sin\phi\cot\theta\frac{\partial}{\partial\phi}|lm\rangle=-\frac{1}{2}[G_{pq}-K_{pq}]. \tag{A·17}$$

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