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X-Ray Scattering from Liquids with Nearly Spherical Molecules*

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For fluids with molecules in which the electron density can be represented by a rapidly converging expansion in spherical harmonics, the intensity of the X-ray scattering can be obtained from a theory by Steele and Pecora. We have re-expanded their result using a different reference frame. This allows us to give a more intuitive physical interpretation of the coefficients of the expansion of the binary correlation function. For any type of molecules, the small angle scattering is given by the compressibility term plus pure orientational contributions. For molecules that are closely spherical, the orientational contribution is of second order in the coefficients of the higher spherical form factors, and therefore small. We have calculated the form factors for NH_3, HF, and H_2O. In all of these molecules, the coefficient of the spherical part is much larger than all the others. This simplifies the interpretation of the experimental data since only three parameters are needed, instead of the six necessary if the orientational part were important.

1. BASIC FORMALISM

The theory of X-ray scattering has been discussed by many authors. For molecules which are not too asymmetric, in the sense that their electron density can be represented by a rapidly converging multipole expansion, a rather general theory has been formulated by Steele and Pecora [1]. We intend to reformulate their result using a different reference frame. Our expansion is somewhat less symmetric than Steele and Pecora's but has a more direct physical and geometrical interpreation. Our procedure relies heavily on the beautiful work of A. ben Reuven and N. D. Gershon [2] for light scattering.

The reference frame is attached to an arbitrarily chosen molecule i. The position of another molecule j is given by a vector $\mathbf{R} \equiv R, \theta, \varphi$ and its orientation by three Euler angles $\mathbf{\Omega} = \alpha, \beta, \gamma$. The two-body correlation function is then of the form $g(\mathbf{R}, \mathbf{\Omega})$.

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Physically, the **R** dependence (which we will call "angular" part) and the **Ω** dependence ("orientational" part) have a different effect on the scattered intensity. The small angle X-ray scattering behavior is given by the orientational part of the correlation function and is quadratic in the coefficients of the nonspherical part of the molecular electron density. The pure angular part however is linear in these coefficients and is relevant to the scattering at large angles.

Following van Hove and Glauber [3], the cross section for elastic scattering can be written as

$$\frac{d\sigma}{d\omega} = \sum_{i,j}^{N} \langle a_i a_j^* e^{i\boldsymbol{\kappa}\cdot\mathbf{R}_{ij}} \rangle, \tag{1.1}$$

where $(d\sigma/d\omega)$ is the differential cross section for the radiation at solid angle ω; $\boldsymbol{\kappa} = \mathbf{k} - \mathbf{k}_0$, where $\mathbf{k}$, $\mathbf{k}_0$ are the wave vectors of the scattered and incident beams. The factors a_i give the amplitude of the radiation scattered by molecule i and finally $\mathbf{R}_{ij} = \mathbf{R}_i - \mathbf{R}_j$ is the center to center distance between the molecules i and j. The brackets indicate the ensemble average of the quantity between them.

The quantities a_i have been discussed by Steele and Pecora [1]. In an arbitrary reference frame **x** we have

$$a_i = Q \int \rho(\mathbf{x})\, e^{i\boldsymbol{\kappa}\cdot\mathbf{x}}\, d\mathbf{x}, \tag{1.2}$$

where $\rho(\mathbf{x})$ is the electron density (or neutron scattering density) and Q is a scattering factor which depends on the nature of the incident radiation. For polarized X-rays,

$$Q = \frac{e}{mc^2}(1 - \sin^2\theta_0 \cos^2\phi_0)^{1/2}. \tag{1.3}$$

For unpolarized X-rays,

$$Q = \frac{e}{mc^2}(1 + \cos^2\theta_0)^{1/2}. \tag{1.4}$$

For neutrons,

$$Q = 1; \qquad \rho(\mathbf{x}) = \sum_{\alpha} S_\alpha \rho_\alpha(\mathbf{x}). \tag{1.5}$$

In these formulas, e is the electron charge, m is the electron mass, c is the velocity of the light in the vacuum, θ_0 is the angel between **k** and $\mathbf{k}_0$ and ϕ_0 is the angle between the displacement vectors of the electric fields for the incident and scattered fields. In the case of neutron scattering X represents the nuclear coordinates and S_α is the neutron cross section for the nucleus α. We will be mostly concerned with X-ray scattering in what follows.

The standard form of Rayleigh's expansion is

$$e^{i\boldsymbol{\kappa}\cdot\mathbf{x}} = \sum_{m=0}^{\infty} i^m(2m+1)[\mathbf{C}^m(\hat{\boldsymbol{\kappa}}) \odot \mathbf{C}^m(\hat{\mathbf{x}})]\, j_m(\kappa x), \tag{1.6}$$

where we used the $\odot$ to indicate tensor scalar product [2, 4],

$$\mathbf{a}^m \odot \mathbf{b}^m = \sum_{\mu=-m}^{m} (-)^{m-\mu} a^m_{-\mu} b_\mu{}^m. \tag{1.7}$$

In (1.6) $j_m(x)$ are the spherical Bessel functions, and

$$C_\mu{}^m(\boldsymbol{\kappa}) = \left(\frac{4\pi}{2m+1}\right)^{1/2} (-i)^m Y^*_{m\mu}(\hat{\boldsymbol{\kappa}}) \tag{1.8}$$

are the standarized spherical harmonics of Fano and Racah [4], and the notation $\hat{\boldsymbol{\kappa}}$ means angular part of vector $\boldsymbol{\kappa}$. $Y_{m\mu}(\hat{\boldsymbol{\kappa}})$ is the usual surface (spherical) harmonic.

Using (1.6) in (1.2) we get

$$\mathbf{a}_i = \sum_{m=0}^{\infty} i^m \mathbf{C}^m(\hat{\boldsymbol{\kappa}}) \odot \mathbf{a}^m(i), \tag{1.9}$$

with

$$a_\mu{}^m(i) = Q(2m+1) \int dr\, d\hat{\mathbf{x}}\, j_m(\kappa r)\, C_\mu{}^m(\hat{x})\, \rho(\mathbf{x})\, r^2. \tag{1.10}$$

Using the well-known theorems for the transformation of irreducible tensorial sets [5] and the fact that $\rho(\mathbf{x})$ is a scalar, we get

$$a_\mu{}^m(i) = \sum_{\mu'=-m}^{m} a^m_{\mu'}(0)\, D^m_{\mu'\mu}(\boldsymbol{\Omega}_i), \tag{1.11}$$

where the nonspherical form factors $a_\mu{}^m(0)$ are now defined in a frame attached to the molecule and $D^m_{\mu'\mu}(\boldsymbol{\Omega})$ are the matrix elements of the rotation operator [5]. $\boldsymbol{\Omega}_i \equiv \alpha_i, \beta_i, \gamma_i$ represent the Euler angles that give the orientation of the molecule i with respect to an arbitrary frame. From (1.10) we see that

$$a^{m*}_\mu = (-)^{m-\mu} a^m_{-\mu}. \tag{1.12}$$

Replacing (1.6) and (1.9) into (1.1) and also using (1.12), we get

$$\frac{d\sigma}{d\omega} = \sum_{m,n,l} (i)^{m+l-n}(2m+1) \sum_{i,j} \langle j_m(\kappa R_{ij})[\mathbf{C}^m(\hat{\boldsymbol{\kappa}}) \odot \mathbf{C}^m(\hat{\mathbf{R}}_{ij})] \times [\mathbf{a}^n(i) \odot \mathbf{C}^n(\hat{\boldsymbol{\kappa}})][\mathbf{a}^l(j) \odot \mathbf{C}^l(\hat{\boldsymbol{\kappa}})]\rangle. \tag{1.13}$$

This expression can be put into a more convenient form using a theorem of tensor algebra [4]

$$\frac{d\sigma}{d\omega} = \sum_{m,n,l,p,q} (i)^{m+l-n}(2m+1) \sum_{i,j} \langle j_m(\kappa R_{ij})\{[\mathbf{a}^n(i) \otimes \mathbf{a}^l(j)]^q \otimes \mathbf{C}^m(\hat{\mathbf{R}}_{ij})\}^p\rangle \odot \{[\mathbf{C}^n(\hat{\boldsymbol{\kappa}}) \otimes \mathbf{C}^l(\hat{\boldsymbol{\kappa}})]^q \otimes \mathbf{C}^m(\hat{\boldsymbol{\kappa}})\}^p. \tag{1.14}$$

Here we have used the definitions of the cross product

$$(\mathbf{a}^l \otimes b^m)^n_\nu = \sum_{\lambda,\mu} (n\nu \mid l\lambda m\mu)\, a_\lambda{}^l b_\mu{}^m, \tag{1.15}$$

where the angular coupling coefficients $(n\nu \mid l\lambda m\mu)$ have been defined in the literature [2, 4, 5]. Equations (1.14) is the scalar product of two tensors, one of which depends only on the direction of $\boldsymbol{\kappa}$, that is, on the geometry of the experiment, while the other depends only molecular parameters. Since we are dealing with isotropics fluids this factor has to be a scalar quantity, which means that only the term with $p = 0$ is different from zero after the averaging over the ensemble has been done. Using

$$(0\,0 \mid l\lambda m\mu) = (-)^{l-\lambda}(2l+1)^{-1/2}\, \delta_{lm}\delta_{\lambda,-\mu}$$

and the definitions of the scalar and direct products, we get after some algebra,

$$\frac{d\sigma}{d\omega} = \sum_{m,n,l} F^m_{nl} \cdot \Phi^m_{nl}\,, \tag{1.17}$$

where

$$\Phi^m_{nl} = (i)^{m+l-n}\{[\mathbf{C}^n(\hat{\boldsymbol{\kappa}}) \otimes \mathbf{C}^l(\hat{\boldsymbol{\kappa}})]^m \odot \mathbf{C}^m(\hat{\boldsymbol{\kappa}})\}, \tag{1.18}$$

$$F^m_{nl} = \sum_{i,j} \langle j_m(\kappa R_{ij})\{[\mathbf{a}^n(i) \otimes \mathbf{a}^l(j)]^m \odot \mathbf{C}^m(\hat{R}_{ij})\}\rangle. \tag{1.19}$$

Using standard tensor algebra we can show in a straight forward manner that

$$\Phi^m_{nl} = \begin{pmatrix} m & n & l \\ 0 & 0 & 0 \end{pmatrix} (2m+1)^{1/2}(-)^n, \tag{1.20}$$

where we have used the standard notation for Wigner's $3j$ symbols.

To compute F^m_{nl}, notice first that since it is a scalar, it does not depend on the choice of coordinate system. We may therefore choose a frame attached to molecule i. Using again the definitions (1.7), (1.11), and (1.15) we get

$$F^m_{nl} = \sum_{\nu,\lambda,\lambda'} (-)^{n-l-m}(2m+1)^{1/2} \begin{pmatrix} m & n & l \\ -\mu & \nu & \lambda \end{pmatrix} a_\nu{}^n(0)\, a^l_{\lambda'}(0) \times \sum_{i,j} \langle j_m(\kappa R_{ij})\, D^l_{\lambda'\lambda}(\mathbf{\Omega}_{ij})\, C^m_{-\mu}(\hat{\mathbf{R}}_{ij})\rangle. \tag{1.21}$$

Replacing (1.20) and (1.21) into (1.17), we get

$$\frac{d\sigma}{d\omega} = \sum_{m,n,l} (-)^n \begin{pmatrix} m & n & l \\ 0 & 0 & 0 \end{pmatrix} \sum_{\nu,\lambda,\lambda'} \begin{pmatrix} m & n & l \\ -\mu & \nu & \lambda \end{pmatrix} a_\nu{}^n(0)\, a^l_{\lambda'}(0) \times (2m+1) \sum_{i,j} \langle j_m(\kappa R_{ij})\, D^l_{\lambda\lambda'}(\mathbf{\Omega}_{ij})\, C^m_{-\mu}(\hat{\mathbf{R}}_{ij})\rangle. \tag{1.22}$$

Performing now the ensemble average, we get (the details follow closely Ref. [1]):

$$\frac{1}{V}\frac{d\sigma}{d\omega} = \rho \Big\{ \sum_{m,\mu} |\, a_\mu{}^m \,|^2 + \frac{\rho}{8\pi^2} \sum_{m,n,l} \sum_{\nu,\lambda,\lambda'} (2m+1)(-)^n \begin{pmatrix} m & n & l \\ 0 & 0 & 0 \end{pmatrix} \times \begin{pmatrix} m & n & l \\ -\mu & \nu & \lambda \end{pmatrix} a_\nu{}^n(0)\, a^l_{\lambda'}(0) \int d\mathbf{R}\, d\mathbf{\Omega}[g(\mathbf{R},\mathbf{\Omega}) - 1]\, j_m(\kappa R) \times D^l_{\lambda'\lambda}(\mathbf{\Omega})\, C^m_{-\mu}(\hat{\mathbf{R}}) \Big\}. \tag{1.23}$$

Let us now discuss this expression. Using the fact that for $\boldsymbol{\kappa} = 2\,|\,\mathbf{k}\,|\sin\frac{1}{2}\theta_0 \to 0$, the asymptotic form of the spherical Bessel functions is

$$j_m(\kappa R) \approx (\kappa R)^m/(2m+1)!!. \tag{1.24}$$

Then, the limiting scattering law for very small angles is given by the term with $m = 0$,

$$\frac{1}{V}\frac{d\sigma}{d\omega} = \rho \Big\{ \sum_{m,\mu} |\, a_\mu{}^m \,|^2 + \left(\frac{\rho}{8\pi^2}\right) |\, a^0_0 \,|^2 \int d\mathbf{R}\, d\mathbf{\Omega}\, j_0(\boldsymbol{\kappa} R) \times [g(\mathbf{R},\mathbf{\Omega}) - 1] + \left(\frac{\rho}{8\pi^2}\right) \sum_{n=0}^{\infty} \sum_{\nu,\lambda'} (-)^{\nu+n} a_\nu{}^n a^n_{\lambda'} \int d\mathbf{R}\, d\mathbf{\Omega}\, g(\mathbf{R},\mathbf{\Omega})\, D^n_{\lambda',-\nu}(\Omega)\, j_0(\kappa r) \Big\}. \tag{1.25}$$

The first term in (1.25) is the "self" term, and is $\boldsymbol{\kappa}$ independent. The second term is the compressibility term and can be obtained from thermodynamic considerations. The third and last term is due to pure orientation correlations. It is wavelength independent if we are in the region where $j_0(\kappa R) \approx 1$ and therefore contributes also to depolarization effects in light scattering. Notice that the correction to the compressibility are of second order in $a_\nu{}^n$, so that in many cases, when $a_0{}^0 \ll a_\nu{}^n$ they will amount to only very small corrections.

When only linear terms in the nonspherical form factors are relevant, then we have to have either $l, \lambda' = 0$ or $n, \nu = 0$.

In the first case we need $m, \mu = n, \nu$ and we get a contribution of the form

$$A_1 = \left(\frac{\rho^2}{8\pi^2}\right) \sum_{\substack{m \neq 0 \\ \mu}} (-)^{m+\mu} a_0{}^0 a_\mu{}^m \times \int d\mathbf{R}\, d\mathbf{\Omega}\, g(\mathbf{R}, \mathbf{\Omega})\, j_m(\kappa R)\, C^m_{-\mu}(\hat{\mathbf{R}}). \quad (1.26)$$

This contribution does not depend on the mutual orientation of the molecules, but does depend on the direction of the vector $\hat{\mathbf{R}}$ in the frame attached to molecule i. We have called this the angular part of the correlation function.

In the second case, when $n, \nu = 0$ we get

$$A_2 = \left(\frac{\rho^2}{8\pi^2}\right) \sum_{m,\mu,\lambda'} (-)^{m+\mu} a_0{}^0 a^m_{\lambda'} \times \int d\mathbf{R}\, d\mathbf{\Omega}\, g(\mathbf{R}, \mathbf{\Omega})\, j_m(\kappa R)\, D^m_{\lambda,\mu}(\mathbf{\Omega})\, C^m_{-\mu}(\hat{\mathbf{R}}). \quad (1.27)$$

If we rotate the reference frame through an angle $-\mathbf{\Omega}$, $\mathbf{R} \to -\mathbf{R}'$ (which is equivalent to taking j as the reference molecule), we get

$$A_2 = \left(\frac{\rho^2}{8\pi^2}\right) a_0{}^0 \sum_{m,\mu} (-)^{m-\mu} a_\mu{}^m \int d\mathbf{R}\, d\mathbf{\Omega}\, g(\mathbf{R}, \mathbf{\Omega})\, C^m_{-\mu}(\hat{\mathbf{R}})\, j_m(\kappa R), \quad (1.28)$$

where now $\hat{\mathbf{R}}'$ are the angular coordinates of the center of molecule i in a frame attached to molecule j. Obviously, both these contributions are equal. We have then

$$\frac{1}{V}\frac{d\sigma}{d\omega} = \rho \left\{\sum |\, a_\mu{}^m\,|^2 + \left(\frac{\rho}{8\pi^2}\right) |\, a_0{}^0\,|^2 \int d\mathbf{R}\, d\mathbf{\Omega}\, j_0(\kappa R)[g(\mathbf{R}, \mathbf{\Omega}) - 1]\right\} + 2A_1\,. \quad (1.29)$$

For nearly spherical molecules, we have only scattering due to the "angular" part of the correlation function. The pure orientational correlations will not contribute to the scattering, so that we need to find only a 3-dimensional correlation function $g(\mathbf{R})$ to interpret the scattering data. Considerations similar to that of Narten and Levy [6] on the number of experiments necessary to fully determine this correlation function apply here. Notice however that a different experiment will lead us to a completely different expansion: Although the X-ray scattering will depend on only a few coefficients because the series is rapidly converging, this will not be the case in a neutron scattering experiment, where we expect a poorly converging series. We hope to discuss these matters in more detail in the future.

2. Form Factor Calculation For Water and Similar Molecules

The nonspherical form factors, defined by Eq. (1.10) can be evaluated from electronic wavefunctions. The procedure is very similar to the one used by Stewart [7] in the calculation of his generalized X-ray scattering factors for structural analysis. Our case is however somewhat different because most of the electron density is centered around one atom (the oxygen) while only a fraction is effectively centered around the hydrogen atoms. This fact was used by Moccia [8] to obtain SCF-MO, one-center expansions for various simple molecules, and water among them [9]. These wavefunctions are quite good when compared with the best SCF available, the calculation of Neumann and Moskowitz [10]. The difference of about 0.17 % in the total energy is due to a great extent to the fact that the one-centered wavefunctions are rather poor representations of the electron density around the hydrogen nucleus, and while these contributions are important for the energy computation, they are much less significant to the X-ray scattering form factors, since only a small part of the electron cloud is contained in them [11].

Quite generally, the electron density is

$$\rho(\mathbf{r}) = 2 \sum_{i=1}^{n} | \Psi_i |^2, \tag{2.1}$$

where n is now the numer of MO's of the molecule and Ψ_i the molecular orbital. In terms of the atomic orbitals we have

$$\Psi_i = \sum_j A_{ij} \varphi_j \,. \tag{2.2}$$

Using the Slater–Moccia [8] orbitals

$$\varphi_i = [(2n_i)!]^{-1/2} \, (2\alpha_i)^{n_i+1/2} \, r^{n_i-1} e^{-\alpha_i r} S_{J_i M_i} \,, \tag{2.3}$$

where $S_{J_iM_i}$ is the tesseral spherical harmonic, and together with the other symbols, has been defined by Moccia [8].

Replacing (2.1), (2.2), and (2.3) into (1.10) we get

$$a_M{}^J = Q(2J+1) \sum_p \sum_{i,j} A_{pi} A_{pj} F(JM \mid J_i M_i J_j M_j) \, H(J, ij), \tag{2.4}$$

with

$$F(JM \mid J_i M_i J_j M_j) = \int d\Omega \; C_M{}^J(\Omega) \, S_{J_i M_i}(\Omega) \, S_{J_j M_j}(\Omega) \tag{2.5}$$

and

$$H(J, ij) = \frac{(2\alpha_i)^{n_i+1/2} (2\alpha_j)^{n_j+1/2}}{[(2n_i)! \, (2n_j)!]^{1/2}} \int dr \; r^{n_i+n_j} j_J(\kappa r) \, e^{-r(\alpha_i+\alpha_j)}. \tag{2.6}$$

TABLE I

Form factors for water from Moccia's [8] orbitals. The units for the form factors are the standard electron units. The reference frame is that of Moccia's paper.

κ^0(Å^{-1})	a_0^0	a_0^1	a_0^2	a_2^2	a_0^3	a_2^3
0.0	10.00	0.00	0.00	0.00	0.00	0.00
0.5	9.78	0.17	0.00	0.00	0.00	0.00
1.0	9.15	0.29	0.00	−0.02	0.00	−0.01
1.5	8.25	0.36	0.01	−0.04	0.00	−0.01
2.0	7.23	0.35	0.01	−0.05	−0.01	−0.02
2.5	6.22	0.30	0.01	−0.06	−0.01	−0.02
3.0	5.30	0.22	0.01	−0.06	−0.02	−0.03
3.5	4.52	0.14	0.01	−0.05	−0.02	−0.03
4.0	3.88	0.08	0.01	−0.04	−0.02	−0.03
4.5	3.37	0.03	0.00	−0.03	−0.02	−0.02
5.0	2.96	0.00		−0.02	−0.02	−0.02
5.5	2.65	−0.02		−0.01	−0.01	−0.02
6.0	2.40	−0.03		0.00	−0.01	−0.01
6.5	2.21	−0.04		0.00	−0.01	−0.01
7.0	2.05	−0.04		0.01	−0.01	−0.01
7.5	1.93	−0.04		0.01		−0.01
8.0	1.83	−0.03		0.01		−0.01
8.5	1.75	−0.03		0.01		
9.0	1.68	−0.02		0.01		
9.5	1.62	−0.02		0.01		
10.0	1.57	−0.01		0.01		
10.5	1.53	−0.01		0.01		
11.0	1.49	−0.01		0.01		
11.5	1.45	−0.01		0.01		
12.0	1.42	0.00				
12.5	1.38	0.00				
13.0	1.35					
13.5	1.32					
14.0	1.28					
14.5	1.25					
15.0	1.22					
15.5	1.19					
16.0	1.16					
16.5	1.13					
17.0	1.10					

TABLE II

Form factors for Ammonia from Moccia's [8] orbitals. The form factors are given in standard electron units and the frame for the molecule is that of Moccia's paper

$\kappa(\text{Å}^{-1})$	a_0^0	a_0^1	a_0^2	a_0^3
0.0	10.00	0.00	0.00	0.00
0.5	9.68	−0.16	−0.01	0.00
1.0	8.82	−0.27	−0.03	0.00
1.5	7.63	−0.32	−0.05	0.01
2.0	6.37	−0.30	−0.07	0.02
2.5	5.23	−0.23	−0.08	0.03
3.0	4.29	−0.16	−0.08	0.04
3.5	3.56	−0.10	−0.07	0.04
4.0	3.01	−0.04	−0.05	0.04
4.5	2.62	−0.01	−0.04	0.03
5.0	2.33	0.01	−0.02	0.03
5.5	2.12	0.02	−0.01	0.02
6.0	1.96	0.03	0.00	0.02
6.5	1.84	0.03	0.00	0.01
7.0	1.75	0.03	0.01	0.01
7.5	1.68	0.02	0.01	0.01
8.0	1.62	0.02	0.01	0.01
8.5	1.56	0.02	0.01	0.01
9.0	1.52	0.01	0.01	
9.5	1.48	0.01	0.01	
10.0	1.44	0.01	0.01	
10.5	1.40		0.01	
11.0	1.37		0.01	
11.5	1.33		0.01	
12.0	1.30		0.01	
12.5	1.26		0.01	
13.0	1.23		0.01	
13.5	1.19		0.01	
14.0	1.15			
14.5	1.12			
15.0	1.08			
15.5	1.05			
16.0	1.01			
16.5	0.98			
17.0	0.95			

TABLE III

Form factors for HF from Moccia's orbitals [8]. The form factors are given in standard electron units and the frame for the molecule is that of Moccia's paper

$\kappa(\text{Å}^{-1})$	a_0^0	a_0^1	a_0^2	a_0^3
0.0	10.00	0.00	0.00	0.00
0.5	9.84	0.11	0.01	0.00
1.0	9.39	0.21	0.02	0.00
1.5	8.72	0.26	0.04	0.00
2.0	7.92	0.27	0.05	0.01
2.5	7.08	0.24	0.07	0.01
3.0	6.25	0.20	0.07	0.02
3.5	5.49	0.15	0.06	0.02
4.0	4.82	0.09	0.05	0.02
4.5	4.24	0.05	0.04	0.02
5.0	3.76	0.02	0.03	0.02
5.5	3.35	−0.01	0.02	0.02
6.0	3.01	−0.02	0.01	0.02
6.5	2.73	−0.03	0.00	0.01
7.0	2.50	−0.03	−0.01	0.01
7.5	2.31	−0.03	−0.01	0.01
8.0	2.15	−0.03	−0.01	0.01
8.5	2.02	−0.03	−0.02	0.01
9.0	1.91	−0.03	−0.02	
9.5	1.82	−0.02	−0.02	
10.0	1.74	−0.02	−0.02	
10.5	1.68	−0.02	−0.02	
11.0	1.62	−0.01	−0.02	
11.5	1.57	−0.01	−0.01	
12.0	1.53	−0.01	−0.01	
12.5	1.49	−0.01	−0.01	
13.0	1.45		−0.01	
13.5	1.42		−0.01	
14.0	1.38		−0.01	
14.5	1.35		−0.01	
15.0	1.32		−0.01	
15.5	1.30		−0.01	
16.0	1.27		−0.01	
16.5	1.24		−0.01	
17.0	1.21			

The integrals (2.5) are simply related to the Clebsch–Gordon coefficients and can be found in tables [5]. The integrals (2.6) can be evaluated analytically [7].

We have computed the form factors for H_2O, NH_3, and HF. The results are shown in the Tables I–III. The results show that the main two contributions arise from the $a_0{}^0$ and $a_0{}^1$ terms. The $a_0{}^1$ term however does not give any deformation of of the spherical shape. It merely represents a shift of the center of the electron cloud. The fact that the $J = 2$ terms are very small means that there is almost no distortion from a spherical shape into an ellipsoidal one. Although this might be true to a certain extent, it could also be due to the use of one-centered MO, which tend to round off the electron distribution. At any rate for the reasons that we discussed above, wo do not expect this to cause a serious error.

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