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The Orientational Average of exp[2πi(h.x + k.y + l.z)]

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The unit vectors $\mathbf{e}_1,\mathbf{e}_2,\mathbf{e}_3$ form a fixed orthogonal coordinate frame. The unit vectors $\mathbf{e}'_1,\mathbf{e}'_2,\mathbf{e}'_3$ form a movable orthogonal frame with the same origin as the fixed frame. All orientations in space of the movable frame are assumed to be equally probable. Under these conditions the average of $\exp[2\pi i(\Sigma^3_{i,j=1} a_{ij}\,\mathbf{e}_i.\mathbf{e}'_j)]$ is calculated. As an application the average of $\exp[2\pi i(\mathbf{h}.\mathbf{x} + \mathbf{k}.\mathbf{y} + \mathbf{l}.\mathbf{z})]$ is calculated where the vectors **h**,**k**,**l** are specified and where the magnitudes of the vectors **x**,**y**,**z** and the angles between them are specified. This integral is of importance in utilizing *a priori* knowledge of molecular structure as an aid in solving the phase problem.

Introduction

The orientational average of $\exp[2\pi i(\mathbf{h}.\mathbf{x} + \mathbf{k}.\mathbf{y})]$ has been derived by Hauptman (1965). This average is of importance in the calculation of the phase of a triplet when use is made of chemical information (Heinerman, 1977; Main, 1976).

The average of $\exp[2\pi i(\mathbf{h}.\mathbf{x} + \mathbf{k}.\mathbf{y} + \mathbf{l}.\mathbf{z})]$ is of importance in the calculation of the phase of a quartet (Heinerman, Krabbendam & Kroon, 1977).

The derivation

In order to obtain the average of $\exp[2\pi i(\mathbf{h}.\mathbf{x} + \mathbf{k}.\mathbf{y} + \mathbf{l}.\mathbf{z})]$ we shall take a fixed orthogonal coordinate frame $\mathbf{e}_1,\mathbf{e}_2,\mathbf{e}_3$ to describe the vectors **h**,**k**,**l** and a movable orthogonal coordinate frame $\mathbf{e}'_1,\mathbf{e}'_2,\mathbf{e}'_3$ with the same origin for the vectors **x**,**y**,**z**.

The position of the frame $\mathbf{e}'_1,\mathbf{e}'_2,\mathbf{e}'_3$ relative to the frame $\mathbf{e}_1,\mathbf{e}_2,\mathbf{e}_3$ can best be described by the well-known Eulerian angles (θ,φ,ψ) defined as:

θ: the angle between $\mathbf{e}'_3$ and $\mathbf{e}_3$ ranging from 0 to π.

φ: the angle from $\mathbf{e}_1$ to the vector $\mathbf{e}_3 \times \mathbf{e}'_3$ measured in an anti-clockwise direction and ranging from 0 to 2π.

ψ: the angle from the vector $\mathbf{e}_3 \times \mathbf{e}'_1$ to $\mathbf{e}'_1$ measured in an anti-clockwise direction and ranging from 0 to 2π.

We then have:

$$\begin{aligned}
\mathbf{e}'_1 &= (\cos\varphi\cos\psi - \sin\varphi\sin\psi\cos\theta)\,\mathbf{e}_1 + (\sin\varphi\cos\psi \\
&\quad + \cos\varphi\sin\psi\cos\theta)\,\mathbf{e}_2 + \sin\psi\sin\theta\mathbf{e}_3 \\
\mathbf{e}'_2 &= (-\cos\varphi\sin\psi - \sin\varphi\cos\psi\cos\theta)\,\mathbf{e}_1 \\
&\quad + (-\sin\varphi\sin\psi + \cos\varphi\cos\psi\cos\theta)\,\mathbf{e}_2 \\
&\quad + \cos\psi\sin\theta\mathbf{e}_3 \\
\mathbf{e}'_3 &= \sin\varphi\sin\theta\mathbf{e}_1 - \cos\varphi\sin\theta\mathbf{e}_2 + \cos\theta\mathbf{e}_3.
\end{aligned} \tag{1}$$

Let $\mathbf{h}_1,\mathbf{k}_1,\mathbf{l}_1,\mathbf{x}_1,\mathbf{y}_1,\mathbf{z}_1$ be the unit vectors in the directions of, respectively, **h**,**k**,**l**,**x**,**y**,**z**. Then:

$$\begin{aligned}
\mathbf{h}_1 &= \alpha_1\mathbf{e}_1 + \alpha_2\mathbf{e}_2 + \alpha_3\mathbf{e}_3 \\
\mathbf{k}_1 &= \beta_1\mathbf{e}_1 + \beta_2\mathbf{e}_2 + \beta_3\mathbf{e}_3 \\
\mathbf{l}_1 &= \gamma_1\mathbf{e}_1 + \gamma_2\mathbf{e}_2 + \gamma_3\mathbf{e}_3 \\
\mathbf{x}_1 &= \alpha'_1\mathbf{e}'_1 + \alpha'_2\mathbf{e}'_2 + \alpha'_3\mathbf{e}'_3 \\
\mathbf{y}_1 &= \beta'_1\mathbf{e}'_1 + \beta'_2\mathbf{e}'_2 + \beta'_3\mathbf{e}'_3 \\
\mathbf{z}_1 &= \gamma'_1\mathbf{e}'_1 + \gamma'_2\mathbf{e}'_2 + \gamma'_3\mathbf{e}'_3.
\end{aligned} \tag{2}$$

And so we have:

$$\mathbf{h}.\mathbf{x} + \mathbf{k}.\mathbf{y} + \mathbf{l}.\mathbf{z} = \textstyle\sum^3_{i,j=1} a_{ij}\mathbf{e}_i.\mathbf{e}'_j$$

where

$$a_{ij} = hx\,\alpha_i\alpha'_j + ky\,\beta_i\beta'_j + lz\,\gamma_i\gamma'_j. \tag{3}$$

It should be noted that one cannot go from (**x**,**y**,**z**) to (−**x**,−**y**,−**z**) by means of a rotation only. Hence if we assume only the lengths of x, y and z and the angles between them to be known, the orientational average will be the real part of the rotational average of $\exp[2\pi i(\mathbf{h}.\mathbf{x} + \mathbf{k}.\mathbf{y} + \mathbf{l}.\mathbf{z})]$. The average $\langle\exp[2\pi i(\mathbf{h}.\mathbf{x} + \mathbf{k}.\mathbf{y} + \mathbf{l}.\mathbf{z})]\rangle$ will then be the real part of:

$$\begin{aligned}
&\frac{1}{8\pi^2}\int\limits_0^{\pi} d\theta\sin\theta\int\limits_0^{2\pi} d\varphi\int\limits_0^{2\pi} d\psi\exp\{2\pi i[a_{11}(\cos\varphi\cos\psi \\
&\quad - \sin\varphi\sin\psi\cos\theta) - a_{12}(\cos\varphi\sin\psi \\
&\quad + \sin\varphi\cos\psi\cos\theta) + a_{13}\sin\varphi\sin\theta + a_{21}(\cos\psi\sin\varphi \\
&\quad + \sin\psi\cos\varphi\cos\theta) + a_{22}(\cos\psi\cos\varphi\cos\theta \\
&\quad - \sin\varphi\sin\psi) - a_{23}\cos\varphi\sin\theta + a_{31}\sin\psi\sin\theta \\
&\quad + a_{32}\cos\psi\sin\theta + a_{33}\cos\theta]\}.
\end{aligned} \tag{4}$$

Let us take the transformation:

$$\begin{aligned}\varphi+\psi&=2u\\ \varphi-\psi&=2v.\end{aligned}\tag{5}$$

This is similar to Hauptman's (1965) transformation. Because of the periodicity we see that we may take u and v ranging from 0 to 2π. After this transformation the integral can be rewritten as:

$$\frac{1}{8\pi^2}\int_0^\pi d\theta\sin\theta\int_0^{2\pi}du\int_0^{2\pi}dv\exp\left\{i\left[A\cos(2u+\alpha)\cos^2\left(\frac{\theta}{2}\right)+B\cos(2v+\beta)\sin^2\left(\frac{\theta}{2}\right)+C\cos(u+v+\gamma)\sin\theta+D\cos(u-v+\delta)\sin\theta+a_{33}\cos\theta\right]\right\},\tag{6}$$

where

$$2\pi[(a_{11}+a_{22})\cos 2u+(a_{21}-a_{12})\sin 2u]=A\cos(2u+\alpha)$$

$$2\pi[(a_{11}-a_{22})\cos 2v+(a_{21}+a_{12})\sin 2v]=B\cos(2v+\beta)$$

$$2\pi[-a_{23}\cos(u+v)+a_{13}\sin(u+v)]=C\cos(u+v+\gamma)$$

$$2\pi[a_{32}\cos(u-v)+a_{31}\sin(u-v)]=D\cos(u-v+\delta),$$

and where A, B, C and D are real, so that, for instance,

$$A=2\pi[(a_{11}+a_{22})^2+(a_{21}-a_{12})^2]^{1/2}.\tag{7}$$

We shall now regard the coefficients a_{ij} in the integral (4) as general real coefficients. These coefficients can be regarded as forming a matrix:

$$(a_{ij})=\begin{pmatrix}a_{11}&a_{12}&a_{13}\\a_{21}&a_{22}&a_{23}\\a_{31}&a_{32}&a_{33}\end{pmatrix}.\tag{8}$$

We shall also consider the quantities z, t and s which are defined as follows:

$$\begin{aligned}z^2&=4\pi^2\sum_{i,j}a_{ij}^2\\ t^2&=16\pi^4\sum_{i,j}A_{ij}^2,\end{aligned}\tag{9}$$

where A_{ij} is the cofactor of a_{ij} (e.g. $A_{11}=a_{22}a_{33}-a_{32}a_{23}$),

$$s=8\pi^3\det(a_{ij}),$$

where $\det(a_{ij})$ is the determinant of the matrix (a_{ij}).

We shall now use the theorem of Lerch (Watson, 1966, p. 382) to evaluate this integral. This theorem states that if $f(r)$ is a continuous function of r when $r>0$ such that $\int_0^\infty\exp(-r^2t)f(r)dr=0$ for all sufficiently large positive values of t then $f(r)$ is identically zero.

We shall first show that the integral (4) is a function of the parameters z, t and s only. In order to do this we shall express the parameters z, t and s as functions of $A, B, C, D, \alpha, \beta, \gamma, \delta$ and a_{33}. We then find:

$$\begin{aligned}z^2&=\frac{(A^2+B^2)}{2}+C^2+D^2+a_{33}^2\\ t^2&=\frac{(A^2-B^2)^2}{16}+\frac{(C^2+D^2)(A^2+B^2)}{4}+\frac{a_{33}^2(A^2+B^2)}{2}+C^2D^2\\&\quad+\frac{[ABC^2\cos(\alpha+\beta-2\gamma)+ABD^2\cos(\alpha-\beta-2\delta)]}{2}\\&\quad-a_{33}CDB\cos(\beta-\gamma+\delta)+a_{33}CDA\cos(\alpha-\gamma-\delta)\\ s&=\frac{a_{33}(A^2-B^2)}{4}+\frac{ACD}{2}\cos(\alpha-\gamma-\delta)+\frac{BCD}{2}\cos(\beta+\delta-\gamma).\end{aligned}\tag{10}$$

We now consider the function:

$$f(r)=2r^3\sin\left(\frac{\theta}{2}\right)\cos\left(\frac{\theta}{2}\right)\exp\left(i\left\{A\cos(2u+\alpha)r^2\cos^2\left(\frac{\theta}{2}\right)+B\cos(2v+\beta)r^2\sin^2\left(\frac{\theta}{2}\right)+2r^2\sin\left(\frac{\theta}{2}\right)\cos\left(\frac{\theta}{2}\right)[C\cos(u+v+\gamma)+D\cos(u-v+\delta)]+a_{33}r^2\left[\cos^2\left(\frac{\theta}{2}\right)-\sin^2\left(\frac{\theta}{2}\right)\right]\right\}\right).\tag{11}$$

Then $(8\pi^2)^{-1}\int_0^\infty\exp(-r^2p)\int_0^\pi d\theta\int_0^{2\pi}du\int_0^{2\pi}dv f(r)dr$ becomes under the transformation $x=r\cos(\theta/2)$, $y=r\sin(\theta/2)$:

$$\begin{aligned}(2\pi^2)^{-1}&\int_0^\infty dx\int_0^\infty dy\int_0^{2\pi}du\int_0^{2\pi}dv\,xy\\&\times\exp[i\{Ax^2\cos(2u+\alpha)+By^2\cos(2v+\beta)\\&+2xy[C\cos(u+v+\gamma)+D\cos(u-v+\delta)]\}\\&-x^2(p-ia_{33})-y^2(p+ia_{33})].\end{aligned}\tag{12}$$

After the transformation $x^2\to x$ and $y^2\to y$ this integral becomes finally:

$$\begin{aligned}(8\pi^2)^{-1}&\int_0^\infty dx\int_0^\infty dy\int_0^{2\pi}du\int_0^{2\pi}dv\exp[i\{Ax\cos(2u+\alpha)\\&+By\cos(2v+\beta)+2\sqrt{x}\sqrt{y}[C\cos(u+v+\gamma)\\&+D\cos(u-v+\delta)]\}-x(p-ia_{33})-y(p+ia_{33})].\end{aligned}\tag{13}$$

Let us first perform the u integration with

$$C\cos(u+v+\gamma)+D\cos(u-v+\delta)=Z\cos(u+\zeta)$$

where

$$Z^2=C^2+D^2+2CD\cos(2v+\gamma-\delta)$$
$$Z\cos\zeta=C\cos(v+\gamma)+D\cos(v-\delta),$$
$$Z\sin\zeta=C\sin(v+\gamma)+D\sin(\delta-v), \qquad (14)$$

(13) becomes:

$$(8\pi^2)^{-1}\int_0^\infty \mathrm{d}x\int_0^\infty \mathrm{d}y\int_0^{2\pi}\mathrm{d}v\exp[-x(p-ia_{33})-y(p+ia_{33})+iBy\cos(2v+\beta)]\times\int_0^{2\pi}\mathrm{d}u\exp[iAx\cos(2u+\alpha)+2i\sqrt{x}\sqrt{y}Z\cos(u+\zeta)]. \qquad (15)$$

Expanding:

$$\exp[iAx\cos(2u+\alpha)]=\sum_{+\infty}^{-\infty} i^nJ_n(Ax)\exp[in(2u+\alpha)]$$

$$\exp[2i\sqrt{x}\sqrt{y}Z\cos(u+\zeta)]=\sum_{+\infty}^{-\infty} i^mJ_m(2Z\sqrt{y}\sqrt{x})\exp[im(u+\zeta)], \qquad (16)$$

and substituting this in (15) we get, after the u integration,

$$(4\pi)^{-1}\int_0^\infty \mathrm{d}y\int_0^{2\pi}\mathrm{d}v\int_0^\infty \mathrm{d}x\exp[-x(p-ia_{33})-y(p+ia_{33})+iBy\cos(2v+\beta)]\sum_{-\infty}^{+\infty}(-i)^nJ_n(Ax)J_{2n}(2Z\sqrt{y}\sqrt{x})\times\exp[in(\alpha-2\zeta)]. \qquad (17)$$

Using formula (A.1) of the Appendix for the x integration, we obtain:

$$(4\pi)^{-1}\int_0^\infty \mathrm{d}y\int_0^{2\pi}\mathrm{d}v\exp\left[-y(p+ia_{33})+iBy\cos(2v+\beta)-\frac{Z^2y(p-ia_{33})}{w}\right]w^{-1/2}\sum_{+\infty}^{-\infty}(-i)^nJ_n\left(\frac{AZ^2y}{w}\right)\times\exp[in(\alpha-2\zeta)], \qquad (18)$$

where $w=(p-ia_{33})^2+A^2$. Now

$$\sum_{+\infty}^{-\infty}(-i)^nJ_n\left(\frac{AZ^2y}{w}\right)\exp[in(\alpha-2\zeta)]=\exp\left[-i\left(\frac{AZ^2y}{w}\right)\cos(\alpha-2\zeta)\right]$$

so that (18) becomes:

$$(4\pi)^{-1}w^{-1/2}\int_0^\infty \mathrm{d}y\int_0^{2\pi}\mathrm{d}v\exp\left[-y(p+ia_{33})+iBy\cos(2v+\beta)-\frac{Z^2y(p-ia_{33})}{w}-i\left(\frac{AZ^2y}{w}\right)\cos(\alpha-2\zeta)\right]. \qquad (19)$$

From the definition of Z and ζ (14) we see that:

$$Z^2\cos 2\zeta=C^2\cos(2v+2\gamma)+D^2\cos(2v-2\delta)+2CD\cos(\gamma+\delta)$$
$$Z^2\sin 2\zeta=C^2\sin(2v+2\gamma)-D^2\sin(2v-2\delta)+2CD\sin(\gamma+\delta)$$
$$Z^2=C^2+D^2+2CD\cos(2v+\gamma-\delta). \qquad (20)$$

Substituting these values in (19) the integral becomes:

$$(4\pi)^{-1}w^{-1/2}\int_0^\infty \mathrm{d}y\exp\left[-y(p+ia_{33})-(C^2+D^2)(p-ia_{33})\frac{y}{w}-2iy\left(\frac{ACD}{w}\right)\times\cos(\gamma+\delta-\alpha)\right]\int_0^\pi \mathrm{d}v\exp\left\{iBy\cos(2v+\beta)-2CD(p-ia_{33})\left(\frac{y}{w}\right)\cos(2v+\gamma-\delta)-i\left(\frac{Ay}{w}\right)\times[C^2\cos(2v+2\gamma-\alpha)+D^2\cos(2v-2\delta+\alpha)]\right\}. \qquad (21)$$

Using formula (A3) of the Appendix for the v integration we obtain:

$$\frac{1}{2\sqrt{w}}\int_0^\infty \mathrm{d}y\exp\left\{-y(p+ia_{33})-\left[\frac{(C^2+D^2)(p-ia_{33})}{w}\right]y-2iy\left(\frac{ACD}{w}\right)\cos(\gamma+\delta-\alpha)\right\}J_0(yR),$$

where

$$R^2=B^2-\frac{4C^2D^2(p-ia_{33})^2}{w^2}+\frac{A^2(C^4+D^4)}{w^2}+4i\left[\frac{BCD(p-ia_{33})}{w}\right]\cos(\beta-\gamma+\delta)-2\left(\frac{ABC^2}{w}\right)\times\cos(\alpha+\beta-2\gamma)-2\left(\frac{ABD^2}{w}\right)\cos(\alpha-\beta-2\delta)-4i\left[\frac{ACD(C^2+D^2)(p-ia_{33})}{w^2}\right]\times\cos(\alpha-\gamma-\delta)+2\left(\frac{ACD}{w}\right)^2\cos 2(\alpha-\gamma-\delta). \qquad (22)$$

Putting

$$a_4=(p+ia_{33})-\frac{(C^2+D^2)(p-ia_{33})}{w}-2i\left(\frac{ACD}{w}\right)\cos(\gamma+\delta-\alpha) \quad (23)$$

we obtain, after carrying out the y integration with formula (A.2) of the Appendix, for the integral (21) (for a sufficiently large positive value of p):

$$\tfrac{1}{2}[w(a_4^2+R^2)]^{-\frac{1}{2}}. \quad (24)$$

This becomes, after some calculation,

$$\tfrac{1}{2}[(p^2+z^2)^2+8ips-4t^2]^{-\frac{1}{2}}. \quad (25)$$

It has thus been proved that the integral (4) only depends on z, t and s. Hence there exists a function $g(r,z,t,s)$ of r such that (for a sufficiently large positive p)

$$\int_0^\infty \exp(-r^2p)\,g(r,z,t,s)\,\mathrm{d}r=\tfrac{1}{2}[(p^2+z^2)^2+8pis-4t^2]^{-\frac{1}{2}}. \quad (26)$$

Hence, it is clear that the integral (4) can be given by:

$$B(z,t,s)=\frac{1}{2\pi i}\int_C[(p^2+z^2)^2+8pis-4t^2]^{-\frac{1}{2}}e^p\,\mathrm{d}p, \quad (27)$$

where C is the straight line from $p_0-i\infty$ to $p_0+i\infty$, with p_0 a sufficiently large positive value. As is usual, this contour can be closed by a large semicircle to the left in the complex p plane.

It is clear that many representations of $B(z,t,s)$ can be given. We shall give a representation by expanding $[(p^2+z^2)^2+8pis-4t^2]^{-\frac{1}{2}}$ in powers of $(8pis-4t^2)/(p^2+z^2)$. We can write (for a sufficiently large value of p):

$$[(p^2+z^2)^2+8pis-4t^2]^{-\frac{1}{2}}=\sum_{n=0}^{\infty}\binom{-\frac{1}{2}}{n}\frac{(8isp-4t^2)^n}{(p^2+z^2)^{2n+1}}. \quad (28)$$

Every term in the series (28) has poles in iz and $-iz$. In order to calculate

$$(2\pi i)^{-1}\int_C[(2isp-t^2)^n/(p^2+z^2)^{2n+1}]\,e^p\,\mathrm{d}p$$

we shall calculate the contribution for the pole in iz (the contribution for the pole in $-iz$ is obtained by reversing the signs). Putting

$$u=p-iz \quad (29)$$

we have (for small values of $|u|$)

$$\frac{e^p}{(p+iz)^{2n+1}}=e^{iz}\sum_{k=0}^{\infty}u^k\sum_{l=0}^{k}\binom{-(2n+1)}{l}\frac{1}{(k-l)!}\times(2iz)^{-(2n+l+1)}. \quad (30)$$

Hence the contribution coming from the pole in iz is

$$(-1)^n\exp(iz)\sum_{m=0}^{n}\binom{n}{m}(-2is)^m(2sz+t^2)^{n-m}\times\sum_{l=0}^{2n-m}\binom{-(2n+1)}{l}\frac{1}{(2n-m-l)!}(2iz)^{-(2n+l+1)}, \quad (31)$$

which can be rewritten as

$$\frac{(-1)^n}{(2n)!}\frac{\exp(iz)}{2z}\left[\frac{(2sz+t^2)}{(4z^2)}\right]^n\times\sum_{m=0}^{n}\frac{n!}{m!(n-m)!}\left[-\frac{2is}{(2sz+t^2)}\right]^m G_m^n(z) \quad (32)$$

with

$$G_m^n(z)=-i\sum_{l=0}^{2n-m}\frac{i^{l+2n}(2z)^{-l}(2n+l)!}{[l!(2n-m-l)!]}. \quad (33)$$

Adding the contribution from the pole in $-iz$ we obtain for $B(z,t,s)$:

$$B(z,t,s)=(2z)^{-1}\sum_{n=0}^{\infty}\frac{1}{(n!)^2}\left\{\left[\frac{(2sz+t^2)}{4z^2}\right]^n f_n(z,t,s)-\left[\frac{(t^2-2sz)}{4z^2}\right]^n f_n(-z,t,s)\right\} \quad (34)$$

with

$$f_n(z,t,s)=\exp(iz)\sum_{m=0}^{n}\frac{n!}{m!(n-m)!}\times\left[\frac{-2is}{(2sz+t^2)}\right]^m G_m^n(z). \quad (35)$$

Let us consider the case $s=0$. Using formula (A.4) of the Appendix we get

$$(2\pi z)^{\frac{1}{2}}J_{2n+\frac{1}{2}}(z)=f_n(z,t,0)-f_n(-z,t,0). \quad (36)$$

Hence we obtain

$$B(z,t,0)=\sqrt{\left(\frac{\pi}{2z}\right)}\sum_{n=0}^{\infty}\frac{1}{(n!)^2}\left(\frac{t}{2z}\right)^{2n}J_{2n+\frac{1}{2}}(z). \quad (37)$$

This formula is essentially Hauptman's (1965) result. We shall now give an explicit expression for the parameters z,t,s in the case where we consider $\langle\exp[2\pi i(\mathbf{h}.\mathbf{x}+\mathbf{k}.\mathbf{y}+\mathbf{l}.\mathbf{z})]\rangle$. Putting the expressions (3) for a_{ij} into (9) we get

$$z=2\pi[h^2x^2+k^2y^2+l^2z^2+2(\mathbf{h}.\mathbf{k})(\mathbf{x}.\mathbf{y})+2(\mathbf{h}.\mathbf{l})(\mathbf{x}.\mathbf{z})+2(\mathbf{k}.\mathbf{l})(\mathbf{y}.\mathbf{z})]^{1/2}$$

$$
\begin{aligned}
t = 4\pi^2\{&(\mathbf{h}\times\mathbf{k})^2(\mathbf{x}\times\mathbf{y})^2 + (\mathbf{h}\times\mathbf{l})^2(\mathbf{x}\times\mathbf{z})^2\\
&+ (\mathbf{k}\times\mathbf{l})^2(\mathbf{y}\times\mathbf{z})^2\\
&+ 2[(\mathbf{h}\times\mathbf{k}).(\mathbf{h}\times\mathbf{l})][(\mathbf{x}\times\mathbf{y}).(\mathbf{x}\times\mathbf{z})]\\
&+ 2[(\mathbf{h}\times\mathbf{k}).(\mathbf{k}\times\mathbf{l})][(\mathbf{x}\times\mathbf{y}).(\mathbf{y}\times\mathbf{z})]\\
&+ 2[(\mathbf{h}\times\mathbf{l}).(\mathbf{k}\times\mathbf{l})][(\mathbf{x}\times\mathbf{z}).(\mathbf{y}\times\mathbf{z})]\}^{1/2}
\end{aligned}
$$

$$
s = 8\pi^3[\mathbf{h}.(\mathbf{k}\times\mathbf{l})][\mathbf{x}.(\mathbf{y}\times\mathbf{z})] \qquad (38)
$$

Concluding remarks

If we assume only that the lengths of **x**,**y**,**z** and the angles between them are given, then we find that the orientational average of $\exp[2\pi i(\mathbf{h}.\mathbf{x} + \mathbf{k}.\mathbf{y} + \mathbf{l}.\mathbf{z})]$ is given by the real part of the function $B(z,t,s)$, where $B(z,t,s)$ is given by formula (34).

We note that $B(z,t,s)$ becomes real if $s = 0$. If $s = 0$, then either **l** is a linear combination of **h** and **k**, or **z** is a linear combination of **x** and **y**. After some calculation one readily sees that $B(z,t,0)$ reduces to Hauptman's (1965) expression.

APPENDIX

The formulas we have used are:

(1)
$$
\int_0^\infty e^{-\beta x} J_\nu(bx) J_{2\nu}(2a\sqrt{x}) = \exp\left[\frac{-a^2\beta}{(\beta^2+b^2)}\right] \times J_\nu\left[\frac{a^2 b}{(\beta^2+b^2)}\right](\beta^2+b^2)^{-\frac{1}{2}} \qquad (A.1)
$$

where $\mathrm{Re}\beta > 0$, $b > 0$, $\mathrm{Re}\nu > -\frac{1}{2}$ (Gradshteyn & Ryzhik, 1965, p. 721).

(2)
$$
\int_0^\infty e^{-\alpha x} J_\nu(\beta x) x^\nu \,\mathrm{d}x = (2\beta)^\nu \Gamma(\nu+\tfrac{1}{2})\pi^{-\frac{1}{2}} \times (\alpha^2+\beta^2)^{-(\nu+\frac{1}{2})}, \qquad (A.2)
$$

where $\mathrm{Re}\nu > -\frac{1}{2}$, $\mathrm{Re}\alpha > |\mathrm{Im}\beta|$ (Gradshteyn & Ryzhik, 1965, p. 712).

(3)
$$
\int_0^{2\pi} \exp\left[i\sum_1^n z_i\cos(u+\alpha_i)\right]\mathrm{d}u = 2\pi J_0(Z), \qquad (A.3)
$$

where $Z^2 = (\sum_1^n z_i e^{i\alpha_i})(\sum_1^n z_i e^{-i\alpha_i})$ and z_i is an arbitrary complex.

Proof. Put $Ze^{i\zeta} = \sum_1^n z_i e^{i\alpha_i}$ and $Ze^{-i\zeta} = \sum_1^n z_i e^{-i\alpha_i}$, then $i\sum_1^n z_i\cos(u+\alpha_i) = iZ\cos(u+\zeta)$. So that by Cauchy's theorem:

$$
\int_0^{2\pi}\exp[iZ\cos(u+\zeta)]\,\mathrm{d}u = \int_0^{2\pi}\exp[iZ\cos u] = 2\pi J_0(Z).
$$

(4)
$$
J_{n+\frac{1}{2}}(z) = (2\pi z)^{-\frac{1}{2}}\left[e^{iz}\sum_{k=0}^n i^{-n+k-1}(2z)^{-k}\frac{(n+k)!}{k!(n-k)!} + e^{-iz}\sum_{k=0}^n(-i)^{-n+k-1}(2z)^{-k}\frac{(n+k)!}{k!(n-k)!}\right], \qquad (A.4)
$$

where $n + 1$ is a natural number (Gradshteyn & Ryzhik, 1965, p. 966).

References

Gradshteyn, I. S. & Ryzhik, I. M. (1965). *Table of Integrals, Series and Products*. New York: Academic Press.

Hauptman, H. (1965). *Z. Kristallogr.* **121**, 1–8.

Heinerman, J. J. L. (1977). *Acta Cryst.* A**33**, 100–106.

Heinerman, J. J. L., Krabbendam, H. & Kroon, J. (1977). *Acta Cryst.* A**33**, 873–878.

Main, P. (1976). *Crystallographic Computing Techniques*, edited by F. R. Ahmed, pp. 97–105. Copenhagen: Munksgaard.

Watson, G. N. (1966). *A Treatise on the Theory of Bessel Functions*. Cambridge Univ. Press.