A Generalized Debye Scattering Formula and the Hankel Transform

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The diffracted intensity of an x-ray or neutron diffraction experiment is expressed as an integral over an atomic position distribution function. A generalized Debye scattering formula results. Since this distribution function is expanded into a series of spherical harmonics, an inverse Hankel transform of the intensity allows the calculation of the expansion coefficients which describe the atomic arrangement completely. The connections between the generalized Debye scattering formula and the original Debye formula as well as the Laue scattering formula are derived.

Key words: X-ray Scattering; Debye Formula; Hankel Transform.

1. Introduction

In a previous article of this journal [1], a generalized Debye scattering formula was presented which gives the diffracted X-ray (or neutron) intensity as a function of structure coefficients. Meanwhile, the theory has been developed further by making the structure coefficients dependent on the interatomic distance. The mathematical structure of the resulting Debye scattering formula allows the calculation of the structure coefficients by a Hankel transform (a generalized Fourier transform) of the diffracted intensity.

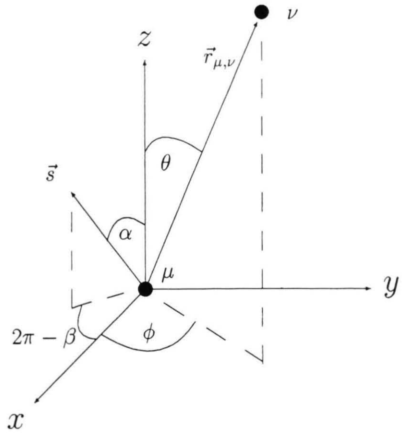

Fig. 1. Vector $\boldsymbol{r}_{\mu,\nu}$ from atom μ to atom ν and scattering vector $\boldsymbol{s}$ with their spherical coordinates.

Consider an assembly of M atoms with fixed positions. Any two of the atoms may be called μ and ν with position vectors $\boldsymbol{r}_\mu$ and $\boldsymbol{r}_\nu$. The difference vector $\boldsymbol{r}_{\mu,\nu} = \boldsymbol{r}_\nu - \boldsymbol{r}_\mu$ will have a certain length r and (using a spherical coordinate system $\mathcal{S}$) a certain direction in $\mathcal{S}$, given by the orientation angles $(\theta_{\mu,\nu}, \phi_{\mu,\nu})$. Now let μ and ν run over all M atoms and observe the direction of $\boldsymbol{r}_{\mu,\nu}$ in space. To $\boldsymbol{r}_{\mu,\nu}$ an orientation distribution function $g_{\mu,\nu} = g_{\mu,\nu}(r, \theta, \phi)$ will correspond[1]. Below, the relation between $g_{\mu,\nu}$ and the diffracted intensity in an X-ray diffraction (or neutron) experiment done on the atomic assembly is outlined.

Another approach to $g_{\mu,\nu}$ is suggested by the usual concept of the crystallographic structure. For a macroscopic crystal, M is large. Nevertheless, its structure is described by a set $\mathcal{E}$ of N atoms[2], where $N \ll M$. In the present notation, two atoms of $\mathcal{E}$ may be called μ and ν again. For any pair $\{\mu, \nu\} \in \mathcal{E}$, the corresponding orientation function $g_{\mu,\nu}$ will be (for the ideal crystal) a δ-function[3] with

$$g_{\mu,\nu}(r, \theta, \phi) = \frac{\delta(r - r_{\mu,\nu})}{r^2_{\mu,\nu}} \frac{\delta(\theta - \theta_{\mu,\nu})}{\sin \theta_{\mu,\nu}} \delta(\phi - \phi_{\mu,\nu}). \quad (1)$$

Any deviation from the ideal crystal structure will change from a δ-function to a more complicated

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[1] $g_{\mu,\nu}$ is actually a distribution density, the distribution itself follows from integrating $g_{\mu,\nu}$ over the entire space.

[2] $\mathcal{E}$ is the set of atoms in the (asymmetric unit of the) elementary cell.

[3] All $g_{\mu,\nu}$ with $\{\mu, \nu\} \in \mathcal{E}$ will be different from another, because crystallographic structures are chosen to be non-redundant. A usual Patterson plot is the graph of all $\boldsymbol{r}_{\mu,\nu}$ with $\{\mu, \nu\} \in \mathcal{E}$.

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function, since then $\boldsymbol{r}_{\mu,\nu}$ will take on a range of orientations. An extreme case is the assumption of an isotropic distribution of $\boldsymbol{r}_{\mu,\nu}$ in space. Then $\boldsymbol{r}_{\mu,\nu}$ covers with constant density the surface of a sphere of radius $r_{\mu,\nu}$. All dependencies on θ and ϕ vanish and one has

$$g_{\mu,\nu}(r,\theta,\phi) = g_{\mu,\nu}(r). \tag{2}$$

In X-ray crystallography, $g_{\mu,\nu}(r)$ is called radial distribution function. If one expects forms of matter with $g_{\mu,\nu}(r,\theta,\phi)$ between the two extreme cases (1) and (2)[4], then one has to find a suitable mathematical expression for $g_{\mu,\nu}(r,\theta,\phi)$. The spherical nature of the problem suggests the use of spherical harmonics $Y_l^m(\theta,\phi)$[5] and the series expansion

$$g_{\mu,\nu}(r,\theta,\phi) = \sum_{l=0}^{L}\sum_{m=-l}^{l} a_{\mu,\nu}^{l,m}(r)Y_l^m(\theta,\phi). \tag{3}$$

A similar expansion without r-dependency has been given earlier [1]. L is the order of expansion. The coefficients $a_{\mu,\nu}^{l,m}(r)$ contain the information about the structure and will be called structure coefficients[6]. Since (3) contains (1) and (2), any structure can be described by (3). For (ideal) crystalline structures, the conventional structure description by the crystal metric and the unit cell composition is surely more convenient. Our motivation for setting up (3) came from considerations on amorphous materials: The traditional assumption of vanishing θ- and ϕ-dependencies in amorphous structures may be too strong. However, this assumption was made in the derivation of the Debye scattering formula for the calculation of the diffracted intensity $i(s)$ in case of gaseous matter. The mathematical structure of the Debye formula allows the Fourier inversion of $i(s)$ in order to yield $g_{\mu,\nu}(r)$. Below, an procedure will be outlined to find $g_{\mu,\nu}(r,\theta,\phi)$ from any atomic assembly.

2. The Generalized Debye Scattering Formula

The intensity $I(s)$ resulting from interferences between atoms μ and ν is the integral over all orientations of $\boldsymbol{r}_{\mu,\nu}$. For any $r_{\mu,\nu}$ = const, an integral over the surface of the sphere with radius $r_{\mu,\nu}$ = const must be carried out. Any possible length $r_{\mu,\nu}$ must be taken into account by integrating over r. A triple integral results. Since all possible pairs $\{\mu,\nu\} \in \mathcal{E}$ contribute to the total intensity, a double sum over μ, ν is necessary. Therefore[7], with α, β, s as the spherical coordinates of the scattering vector $\boldsymbol{s}$ and $f_\mu(s)$, $f_\nu^*(s)$ as (complex conjugated) atomic form factors of atoms μ and ν,

$$\begin{aligned} I(\alpha,\beta,s) = \sum_{\mu=1}^{N}\sum_{\nu=1}^{N} f_\mu f_\nu^* \int_{r=0}^{\infty}\int_{\theta=0}^{\pi}\int_{\phi=0}^{2\pi} g_{\mu,\nu}(r,\theta,\phi) \\ \cdot \exp(2\pi i\,\boldsymbol{r}_{\mu,\nu}\cdot\boldsymbol{s}_{\alpha,\beta})r^2\sin\theta\mathrm{d}\phi\mathrm{d}\theta\mathrm{d}r. \end{aligned} \tag{4}$$

Inserting (3) into (4) and expanding the exponential function into a series of spherical harmonics according to

$$\begin{aligned} \exp(2\pi\imath\boldsymbol{r}\cdot\boldsymbol{s}) = 4\pi\sum_{l=0}^{\infty}\sum_{m=-l}^{l} \imath^l j_l(2\pi r\, s) \\ \cdot Y_l^{m\,*}(\phi,\theta)Y_l^m(\alpha,\beta), \end{aligned} \tag{5}$$

where j_l is the spherical Bessel function, gives

$$\begin{aligned} I(\alpha,\beta,s) = 4\pi\sum_{\mu=1}^{N}\sum_{\nu=1,\neq\mu}^{N} f_\mu f_\nu^* \int_{r=0}^{\infty}\int_{\theta=0}^{\pi}\int_{\phi=0}^{2\pi}\sum_{l=0}^{L}\sum_{m=-l}^{l} \\ a_{\mu,\nu}^{l,m}(r)Y_l^m(\theta,\phi)\sum_{l'=0}^{L}\sum_{m'=-l'}^{l'} \imath^{l'} j_{l'}(2\pi rs)Y_{l'}^{m'\,*}(\theta,\phi) \\ \cdot\ Y_{l'}^{m'}(\alpha,\beta)r^2\sin\theta\mathrm{d}\theta\mathrm{d}\phi\mathrm{d}r + \sum_{\mu=1}^{N} f_\mu^2. \end{aligned} \tag{6}$$

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[4] In the framework of the kinematical theory of X-ray diffraction, one arrives from assumption (1) at the Laue scattering formula and from (2) at the Debye scattering formula.

[5] For the spherical harmonics $Y_l^m(\theta,\phi)$ we use the real–valued form (with P_l^m as the associated Legendre function)

$$Y_l^m(\theta,\phi) = \begin{cases} \sqrt{\frac{2l+1}{4\pi}\frac{(l-m)!}{(l+m)!}}P_l^m(\cos(\theta))\cos(m\phi) \ \text{for}\ m \geq 0, \\ \sqrt{\frac{2l+1}{4\pi}\frac{(l-||m||)!}{(l+||m||)!}}P_l^{||m||}(\cos(\theta))\sin(||m||\phi) \\ \qquad\qquad\qquad\qquad \text{for}\ m < 0. \end{cases}$$

[6] In the Landau theory on phase transitions or in the theory of the structure of metallic glasses [2] similar coefficients are called order parameters.

[7] The triple integral in (4) must be normalized by the normalization factor $Z(\mu,\nu) = \int_{r=0}^{\infty}\int_{\theta=0}^{\pi}\int_{\phi=0}^{2\pi} g_{\mu,\nu}(r,\theta,\phi)r^2\sin\theta\mathrm{d}\phi d\theta\mathrm{d}r$ as in [1]. We omit $Z(\mu,\nu)$ for convenience, but it should be kept in mind when inspecting the dimensions of the equations.

The orthogonality relation[8] between the spherical harmonics considerably reduce the number of terms. A further reduction in the summations over μ and ν comes from the symmetry $a_{\mu,\nu}^{l,m}(r) = (-1)^l a_{\nu,\mu}^{l,m}(r)$. Then the summation over ν needs to run from $\mu + 1$ to N only, since all coefficients $a_{\nu,\mu}^{l,m}(r)$ are accounted for by $2a_{\mu,\nu}^{l,m}(r)$. Finally one receives (similar to the derivation given in [1]) the generalized Debye scattering formula in explicit form

$$I(\alpha,\beta,s) = 8\pi \sum_{\mu=1}^{N-1} \sum_{\nu=\mu+1}^{N} f_\mu f_\nu^* \sum_{\substack{l=0\\ \Delta l=2}}^{L} \sum_{m=-l}^{l} \int_{r=0}^{\infty} a_{\mu,\nu}^{l,m}(r) \imath^l j_l(2\pi r s) r^2 \mathrm{d}r \, Y_l^m(\alpha,\beta) + \sum_{\mu=1}^{N} f_\mu^2. \tag{7}$$

Although (7) contains the imaginary unit $\imath$, the resulting intensity is a real quantity, because the l-summation runs over even l only. Summation terms with odd l cancel each other which is a mathematical expression of inversion symmetry.

The occurrence of the spherical Bessel function j_l of order l with the product rs in its argument suggests to try a inverse Hankel transform[9] of (7) in order to calculate the structure coefficients $a_{\mu,\nu}^{l,m}(r)$ from the measured intensity $i(s)$. To that purpose, the s–dependency of the atomic structure factors $f_\mu(s)$ has to be removed. As described by Warren [5] for the Fourier transform, the function $f_\mu(s)$ is replaced by the constant quantity K_μ. Furthermore, as usual in the analysis of diffraction data, one goes over from $I(s)$ to $i(s)$, the interference function, according to

$$i(\alpha,\beta,s) = I(\alpha,\beta,s) - \sum_{\mu=1}^{N} f_\mu(s)^2. \tag{8}$$

Finally, the use of $k = 2\pi s$ makes the following formulas more symmetrical. After inserting $(kr)^{1/2}/(kr)^{1/2}$ and using $j_l(kr) = \sqrt{\frac{\pi}{2kr}} J_{l+1/2}(kr)$ (where $J_{l+1/2}$ is the Bessel function of fractional order $l + 1/2$) the generalized Debye scattering formula can be written as

$$i(\alpha,\beta,k)k = \sqrt{32\pi^3} \sum_{\mu=1}^{N-1} \sum_{\nu=\mu+1}^{N} K_\mu K_\nu^* \cdot \sum_{\substack{l=0\\ \Delta l=2}}^{L} \sum_{m=-l}^{l} \imath^l H_{\mu,\nu}^{l,m}(k) Y_l^m(\alpha,\beta), \tag{9}$$

with the Hankel transform (the term $a_{\mu,\nu}^{l,m}(r)r$ undergoes the Hankel transform)

$$H_{\mu,\nu}^{l,m}(k) = \int_{r=0}^{\infty} a_{\mu,\nu}^{l,m}(r) r \sqrt{kr} J_{l+1/2}(kr) \mathrm{d}r. \tag{10}$$

3. The Determination of the Patterson Function

According to (10), the structure coefficients $a_{\mu,\nu}^{l,m}(r)$ are related to the intensity coefficients $H_{\mu,\nu}^{l,m}(k)$ by a Hankel transform. Therefore the $H_{\mu,\nu}^{l,m}(k)$ may be called Hankel transformed structure coefficients. The inverse Hankel transform yields the $a_{\mu,\nu}^{l,m}(r)$ by[10]

$$a_{\mu,\nu}^{l,m}(r) r = \int_{k=0}^{\infty} H_{\mu,\nu}^{l,m}(k) \sqrt{kr} J_{l+1/2}(kr) \, \mathrm{d}k. \tag{11}$$

In order to apply (11), one could try to determine the functions $H_{\mu,\nu}^{l,m}(k)$ (as functions of k) from the measured $i(\alpha,\beta,k)$. The number of unknown functions $H_{\mu,\nu}^{l,m}(k)$ is $U = U_{\mu,\nu} U_{l,m} = ((N^2 - N)/2)(L + 1)(L/2 + 1)$ and they can not be recovered from the measured intensity. Carrying out formally the summation over μ and ν in (9) the intensity reads as

$$i(\alpha,\beta,k)k = \sqrt{32\pi^3} \sum_{\substack{l=0\\ \Delta l=2}}^{L} \sum_{m=-l}^{l} \imath^l \overline{H}_l^m(k) Y_l^m(\alpha,\beta), \tag{12}$$

with the Hankel transform

$$\overline{H}_l^m(k) = \sum_{\mu=1}^{N-1} \sum_{\nu=\mu+1}^{N} K_\mu K_\nu^* H_{\mu,\nu}^{l,m}(k). \tag{13}$$

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[8] The orthonormality and completeness relations for the spherical harmonics are $\int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} Y_l^{m*}(\theta,\phi) Y_{l'}^{m'}(\theta,\phi) \sin\theta \mathrm{d}\theta \mathrm{d}\phi = \delta_{mm'}\delta_{ll'}$ and $\sum_{l=0}^{\infty} \sum_{m=-l}^{l} Y_l^{m*}(\theta,\phi) Y_l^m(\theta',\phi') = \frac{\delta(\theta-\theta')}{\sin\theta} \delta(\phi-\phi')$.

[9] The Hankel transform $\mathcal{H}_n(f(x),y)$ of function $f(x)$ for order n and parameter y is defined (see [3, 4]) as $\mathcal{H}_n(f(x),y) = \int_{x=0}^{\infty} f(x)\sqrt{xy} J_n(xy) \mathrm{d}x$.

[10] Normally, $H_{\mu,\nu}^{l,m}(k)$ or $a_{\mu,\nu}^{l,m}(r)$ will not be given analytically, but in form of a discrete data table. Then the Hankel transform must be carried out numerically. A suitable computer program is described in [6].

Then the number of unknowns reduces to $U = U_{l,m} = (L+1)(L/2+1)$. From (12) an equation system can be achieved by variation of α and β at fixed $k = k_i$. This (α, β)–variation provides enough linear independent equations to determine the $\overline{H}_l^m(k)$ at position $k = k_i$, because the rank r_{eqs} of the resulting equation system is equal to $U_{l,m}$. At each $k = k_i$ such an equation system has to be set up up and solved to get the k-dependency of $\overline{H}_l^m(k)$. For $l = 0$ one has $Y_0^0 = 1/\sqrt{4\pi}$ and therefore no dependency on α and β, but then $U = U_{l,m} = 1$ and still $r_{\text{eqs}} = U$.

Once the $\overline{H}_l^m(k)$ have been determined from $i(\alpha, \beta, k)$, then an inverse Hankel transform

$$\overline{a}_l^m(r)r = \int_{k=0}^{\infty} \sqrt{kr}\,\overline{H}_l^m(k) J_{l+1/2}(kr)\mathrm{d}k \tag{14}$$

gives the so-called [1] x-ray structure coefficients $\overline{a}_l^m(r)$. From this relation the name Hankel transformed x-ray structure coefficients follows for the $\overline{H}_l^m(k)$. The x-ray structure coefficients are connected to the structure coefficients by

$$\overline{a}_l^m(r) = \sum_{\mu=1}^{N-1} \sum_{\nu=\mu+1}^{N} K_\mu K_\nu^* a_{\mu,\nu}^{l,m}(r). \tag{15}$$

Suppose, an arbitrary assembly $1 \ldots, \mu, \ldots, \nu, \ldots, N$ of (point-like) atoms and the coordinates $(r_{\mu,\nu}, \theta_{\mu,\nu}, \phi_{\mu,\nu})$ of the corresponding distance vectors $\boldsymbol{r}_{\mu,\nu}$ are given. Referring to (1) and (3), and using the orthogonality of the spherical harmonics, then the coefficients $a_{\mu,\nu}^{l,m}(r)$ can be calculated from the atomic coordinates by the formula

$$a_{\mu,\nu}^{l,m}(r) = \frac{\delta(r - r_{\mu,\nu})}{r_{\mu,\nu}^2} \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} Y_l^{m*}(\theta, \phi) \frac{\delta(\theta - \theta_{\mu,\nu})}{\sin\theta_{\mu,\nu}} \tag{16}$$

$$\cdot\, \delta(\phi - \phi_{\mu,\nu}) \sin\theta \mathrm{d}\phi \mathrm{d}\theta$$

$$= \frac{\delta(r - r_{\mu,\nu})}{r_{\mu,\nu}^2} Y_l^{m*}(\theta_{\mu,\nu}, \phi_{\mu,\nu}). \tag{17}$$

Inserting (17) into (15) and use of the completeness relation of the spherical harmonics[8] makes clear the importance of the x-ray structure coefficients, since

$$\sum_{l=0}^{L} \sum_{m=-l}^{l} \overline{a}_l^m(r) Y_l^m(\theta, \phi) = \sum_{\mu=1}^{N-1} \sum_{\nu=\mu+1}^{N} K_\mu K_\nu^*$$

$$\cdot \sum_{l=0}^{L} \sum_{m=-l}^{l} Y_l^m(\theta, \phi) Y_l^{m*}(\theta_{\mu,\nu}, \phi_{\mu,\nu}) \frac{\delta(r - r_{\mu,\nu})}{r_{\mu,\nu}^2}$$

$$= \sum_{\mu=1}^{N-1} \sum_{\nu=\mu+1}^{N} K_\mu K_\nu^* \frac{\delta(\theta - \theta_{\mu,\nu})}{\sin\theta_{\mu,\nu}} \tag{18}$$

$$\cdot\, \delta(\phi - \phi_{\mu,\nu}) \frac{\delta(r - r_{\mu,\nu})}{r_{\mu,\nu}^2}.$$

Equation (18) is the Patterson function $g(\theta, \phi, r)$ in terms of x-ray structure coefficients. Thus, the Patterson function has been found from (step 1) determining the Hankel transformed structure coefficients $\overline{H}_l^m(k)$ and (step 2) calculating the x-ray structure coefficients $\overline{a}_l^m(r)$. Then one can try (step 3) to recover the structure, that are the position vectors $\boldsymbol{r}_\mu$ from the difference vectors $\boldsymbol{r}_{\mu,\nu}$. Step 3, the interpretation of the Patterson function is the common problem of any x-ray crystallography. Taking into account that the structures treated by the generalized Debye scattering function are (usually) not crystalline, new methods for the interpretation of (18) will be neccessary.

4. Connection to the Debye and Laue Scattering Formulas

4.1. Summation Over Plane Waves

In the kinematical theory of X-ray diffraction, the X-rays are treated as plane waves. The connection between (7) or equivalently (9) and the plane wave formulation is found by inserting (17) into (7)

$$i(\alpha, \beta, k) = 8\pi \sum_{\mu=1}^{N-1} \sum_{\nu=\mu+1}^{N} K_\mu K_\nu^* \sum_{\substack{l=0 \\ \Delta l=2}}^{L} \sum_{m=-l}^{l} \int_{r=0}^{\infty} \mathrm{d}r$$

$$\cdot \frac{\delta(r - r_{\mu,\nu})}{r_{\mu,\nu}^2} \imath^l j_l(kr) r^2\, Y_l^{m*}(\theta_{\mu,\nu}, \phi_{\mu,\nu}) Y_l^m(\alpha, \beta)$$

$$= 8\pi \sum_{\mu=1}^{N-1} \sum_{\nu=\mu+1}^{N} K_\mu K_\nu^* \sum_{\substack{l=0 \\ \Delta l=2}}^{L} \sum_{m=-l}^{l} \imath^l j_l(kr_{\mu,\nu}) \tag{19}$$

$$\cdot\, Y_l^{m*}(\theta_{\mu,\nu}, \phi_{\mu,\nu}) Y_l^m(\alpha, \beta).$$

If the double is rearranged to run over all atoms including the case $\mu = \nu$, then one can change back from $i(k)$ to $I(k)$. In order to arrive at the mathematical expression of plane waves, namely the exponential function, the summation over index l must include the terms for odd l, these are the terms $\sin kr_{\mu,\nu}$. This inclusion does not change the sum, since the sin-terms cancel each other because of the relation $\boldsymbol{r}_{\nu,\mu} = -\boldsymbol{r}_{\mu,\nu}$. Then $I(k)$ becomes

$$I(\alpha,\beta,k) = 4\pi \sum_{\mu=1}^{N}\sum_{\nu=1}^{N} K_\mu K_\nu^* \sum_{l=0}^{L}\sum_{m=-l}^{l} \imath^l j_l(kr_{\mu,\nu}) \cdot Y_l^{m*}(\theta_{\mu,\nu},\phi_{\mu,\nu}) Y_l^m(\alpha,\beta) \quad (20)$$

$$= \sum_{\mu=1}^{N}\sum_{\nu=1}^{N} K_\mu K_\nu^* \exp(\imath \boldsymbol{k}(\alpha,\beta)\cdot \boldsymbol{r}_{\mu,\nu}). \quad (21)$$

Equation (21) is the most general expression for diffracted intensity according to the kinematical theory [7, 5]. All formulas of the kinematical theory are derived from this starting point, in particular the scattering formulas according to Debye and to Laue. Taking the generalized Debye scattering formula (9) as starting point, these formulas can be derived also, as demonstrated below.

4.2. The Debye Equation

The Debye scattering formula [8] follows from (9) for expansion order $L = 0$. Then one has $Y_0^0 = 1/\sqrt{4\pi}$ and $a_{\mu,\nu}^{0,0}(r) = (\delta(r - r_{\mu,\nu})/r_{\mu,\nu}^2)(1/\sqrt{4\pi})$. Inserting gives

$$i(k)k = \sqrt{32\pi^3} \sum_{\mu=1}^{N-1}\sum_{\nu=\mu+1}^{N} K_\mu K_\nu^* \cdot \int_{r=0}^{\infty} a_{\mu,\nu}^{0,0}(r) r\sqrt{kr} J_{1/2}(kr) Y_0^0(\alpha,\beta)\mathrm{d}r$$

$$= \sqrt{32\pi^3} \sum_{\mu=1}^{N-1}\sum_{\nu=\mu+1}^{N} K_\mu K_\nu^* \quad (22)$$

$$\cdot \int_{r=0}^{\infty} \frac{\delta(r - r_{\mu,\nu})}{r_{\mu,\nu}^2} \frac{1}{\sqrt{4\pi}} r\sqrt{kr} J_{1/2}(kr)\mathrm{d}r \frac{1}{\sqrt{4\pi}}.$$

Going back from the Bessel function to the spherical Bessel function j_l with $j_0 = \sin(x)/x$ one has

$$i(k) = 2 \sum_{\mu=1}^{N-1}\sum_{\nu=\mu+1}^{N} K_\mu K_\nu^* \int_{r=0}^{\infty} \frac{\delta(r - r_{\mu,\nu})}{r_{\mu,\nu}^2} \sqrt{\frac{\pi}{2}} \cdot \frac{r}{k}\sqrt{kr}\frac{\sqrt{kr}}{\sqrt{kr}} J_{1/2}(kr)\mathrm{d}r$$

$$= 2 \sum_{\mu=1}^{N-1}\sum_{\nu=\mu+1}^{N} K_\mu K_\nu^* \int_{r=0}^{\infty} \delta(r - r_{\mu,\nu}) j_0(kr)\mathrm{d}r$$

$$= 2 \sum_{\mu=1}^{N-1}\sum_{\nu=\mu+1}^{N} K_\mu K_\nu^* \frac{\sin kr_{\mu,\nu}}{kr_{\mu,\nu}}. \quad (23)$$

Replacing K_μ, K_ν^* by $f_\mu(k)$, $f_\nu^*(k)$ and letting both sums over μ, ν run from 0 to N (with $\mu \neq \nu$) results in the Debye formula.

4.3. The Laue Equation

The Laue scattering formula [7] follows from (9) by using the coefficients $a_{\mu,\nu}^{l,m}(r)$ according to the translational symmetry of a crystal. For simplicity, a simple cubic crystal (no centering) is considered which is composed of a single chemical element, so that $f_\mu f_\nu^* = f^2$. Without loss of generality, one may put the origin of the coordinate system S into atom $\mu = 1$ and then it is sufficient to consider the neighboring atoms ν with coordinates $(r_{\mu,\nu}, \theta = \pi/2, \phi = 0)$,

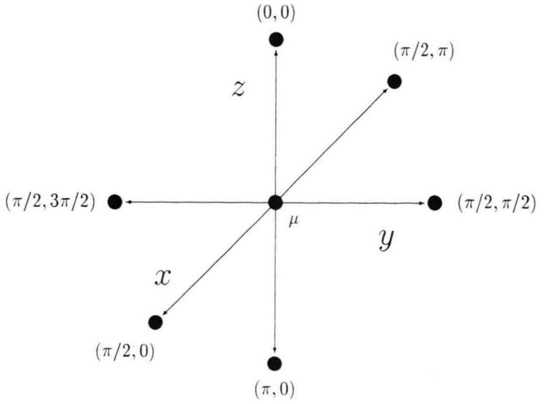

Fig. 2. Atom μ and its six nearest neighbours with their spherical coordinates $(\theta_{\mu,\nu}, \phi_{\mu,\nu}$ in a simple cubic crystal.

$(r_{\mu,\nu}, \pi/2, \pi/2)$, and $(r_{\mu,\nu}, 0, 0)$[11]. In (20) only even values of index l appear, therefore the double summation $\sum\limits_{l} \sum\limits_{m}$ actually returns instead of the exponential function its cosine part only

$$i(\alpha, \beta, s) = 2f^2 \sum_{\mu=1}^{N-1} \sum_{\nu=\mu+1}^{N} \cos(2\pi \boldsymbol{r}_{\mu,\nu} \cdot \boldsymbol{s}). \quad (24)$$

The scalar product $\boldsymbol{r}_{\mu,\nu} \cdot \boldsymbol{s}$ can be evaluated further, since the coordinates of $\boldsymbol{r}_{\mu,\nu}$ are known. In Cartesian coordinates with basis vectors $\boldsymbol{e}_x$, $\boldsymbol{e}_y$, $\boldsymbol{e}_z$ one has $\boldsymbol{r}_{\mu,\nu} = r_{\mu,\nu} \sin\theta \cos\phi \boldsymbol{e}_x + r_{\mu,\nu} \sin\theta \sin\phi \boldsymbol{e}_y + r_{\mu,\nu} \cos\theta \boldsymbol{e}_z$. Furthermore, since a cubic lattice is considered, $r_{\mu,\nu} = (\nu - \mu)a$. Inserting the coordinates $(r_{\mu,\nu}, \theta = \pi/2, \phi = 0)$, $(r_{\mu,\nu}, \pi/2, \pi/2)$, and $(r_{\mu,\nu}, 0, 0)$ yields

$$\begin{aligned} i(\alpha, \beta, s) = 2f^2 \sum_{\mu=1}^{N-1} \sum_{\nu=\mu+1}^{N} & \cos(2\pi((\nu - \mu)a \sin\theta_{\mu,\nu} \\ & \cdot \cos\phi_{\mu,\nu} \boldsymbol{e}_x \cdot \boldsymbol{s} + (\nu - \mu)a \sin\theta_{\mu,\nu} \sin\phi_{\mu,\nu} \boldsymbol{e}_y \cdot \boldsymbol{s} \\ & + (\nu - \mu)a \cos\theta_{\mu,\nu} \boldsymbol{e}_z \cdot \boldsymbol{s})). \end{aligned} \quad (25)$$

At first sight, the trigonometric theorem $\cos(a+b+c) = \cos a \cos b \cos c - \sin a \sin b \cos c - \sin a \cos b \sin c - \cos a \sin b \sin c$ seems to complicate the cos-term. After applying the theorem and inserting the above listed coordinates one sees that all products on the right side containing a sin-term vanish. E.g. for $\mu = 1$, $\nu = 1$ (which corresponds to the x-direction) with $\theta_{\mu=1,\nu=1} = \pi/2, \phi_{\mu=1,\nu=1} = 0$ the cos-argument becomes

$$\begin{aligned} & 2\pi((\nu - \mu)a \sin\theta_{\mu,\nu} \cos\phi_{\mu,\nu} \boldsymbol{e}_x \cdot \boldsymbol{s} + (\nu - \mu)a \\ & \quad \cdot \sin\theta_{\mu,\nu} \sin\phi_{\mu,\nu} \boldsymbol{e}_y \cdot \boldsymbol{s} + (\nu - \mu)a \cos\theta_{\mu,\nu} \boldsymbol{e}_z \cdot \boldsymbol{s})) \\ & = 2\pi((\nu - \mu)a \sin\pi/2 \cos 0 \boldsymbol{e}_x \cdot \boldsymbol{s} + (\nu - \mu)a \\ & \quad \cdot \sin\pi/2 \sin 0 \boldsymbol{e}_y \cdot \boldsymbol{s} + (\nu - \mu)a \cos\pi/2 \boldsymbol{e}_z \cdot \boldsymbol{s})) \\ & = 2\pi((\nu - \mu)a \boldsymbol{e}_x \cdot \boldsymbol{s}). \end{aligned} \quad (26)$$

Therefore, after application of the mentioned trigonometric theorem, (25) may be written as

$$\begin{aligned} i(\alpha, \beta, s) = 2f^2 & \sum_{\mu=1}^{N-1} \sum_{\nu=\mu+1}^{N} \cos 2\pi((\nu - \mu)a \boldsymbol{e}_x \cdot \boldsymbol{s}) \\ & \cdot \sum_{\mu=1}^{N-1} \sum_{\nu=\mu+1}^{N} \cos 2\pi((\nu - \mu)a \boldsymbol{e}_y \cdot \boldsymbol{s}) \\ & \cdot \sum_{\mu=1}^{N-1} \sum_{\nu=\mu+1}^{N} \cos 2\pi((\nu - \mu)a \boldsymbol{e}_z \cdot \boldsymbol{s}). \end{aligned} \quad (27)$$

If the double summation is resequenced in a first step from $\mu = 1 \ldots N-1$, $\nu = \mu+1 \ldots N$ to $\mu = 1 \ldots N$, $\nu = 1 \ldots N$ (with the condition $\mu \neq \nu$) and in a second step to $\mu = -N' \ldots N'$, $\nu = -N' \ldots N'$ (with $N' = N - 1$) then the factor 2 in (27) is absorbed and the sums in (27) can be evaluated by use of $\sum\limits_{n=-p}^{p} \cos(2\pi nax) = \sin((2p+1)\pi ax)/\sin(\pi ax)$ and finally Laue's scattering formula is obtained:

$$\begin{aligned} i(\boldsymbol{s}) = f^2 & \frac{\sin^2 N\pi a \boldsymbol{e}_x \cdot \boldsymbol{s}}{\sin^2 \pi a \boldsymbol{e}_x \cdot \boldsymbol{s}} \frac{\sin^2 N\pi a \boldsymbol{e}_y \cdot \boldsymbol{s}}{\sin^2 \pi a \boldsymbol{e}_y \cdot \boldsymbol{s}} \\ & \cdot \frac{\sin^2 N\pi a \boldsymbol{e}_z \cdot \boldsymbol{s}}{\sin^2 \pi a \boldsymbol{e}_z \cdot \boldsymbol{s}}. \end{aligned} \quad (28)$$

From the sin-quotients the three Laue equations $a\boldsymbol{e}_x \cdot \boldsymbol{s} = h\lambda$ etc. for the occurrence of an (hkl)-reflection are derived in the usual manner.

5. Conclusion

The scattering formula (9) equivalent to the well–known scattering formula (21) of the kinematical theory of X-ray diffraction has been derived. In this new approach the structure is expressed by so–called structure coefficients which can be (in principle) found from a Hankel inversion of the diffracted intensity similar to the well known Fourier inversion[12]. The formula could find applications in amorphous materials or in materials with orientational (dis)order.

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[11] These atoms are the first neighbors of μ along the Cartesian coordinate axes x, y, z.

[12] *Note added in proof:* The integral over r in (7) has the form of a Fourier-Bessel transform [4] which is a special case of the Hankel transform. The above outlined theory can be equivalently formulated in terms of the Fourier-Bessel transform. The difference is the use of $j_l(kr)$ instead of $J_{l+1/2}(kr)$ in the transform pairs (10), (11).

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