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Incommensurate crystallography without additional dimensions

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It is shown that the Euclidean group of translations, when treated as a Lie group, generates translations not only in Euclidean space but on any space, curved or not. Translations are then not necessarily vectors (straight lines); they can be any curve compatible with the parameterization of the considered space. In particular, attention is drawn to the fact that one and only one finite and free module of the Lie algebra of the group of translations can generate both modulated and non-modulated lattices, the modulated character being given only by the parameterization of the space in which the lattice is generated. Moreover, it is shown that the diffraction pattern of a structure is directly linked to the action of that free and finite module. In the Fourier transform of a whole structure, the Fourier transform of the electron density of one unit cell (*i.e.* the structure factor) appears concretely, whether the structure is modulated or not. Thus, there exists a neat separation: the geometrical aspect on the one hand and the action of the group on the other, without requiring additional dimensions.

1. Introduction

A translation in Euclidean space [considered as the point space, mentioned in Wondratschek (2002)] is described by a vector; its action on a point consists in adding one real number to each of its coordinates. However, this does not hold for any space; the ship on a point of the sea can never be considered as translated from another point by a vector, as the direction of sailing changes at any point of the Earth (the sphere); moreover, such a vector would not belong to the surface of the sphere representing our planet. Notwithstanding this, a passenger of the ship would have the impression of a movement of translation.

The concept of translation might then be introduced in any continuous space; there is no reason to limit it to the Euclidean one. The question is how to formalize it. Defining a translation through a vector is not suitable, since such objects are not appropriate in the frame of curved spaces (such as a sphere, a torus or a paraboloid, for instance). As mentioned previously, a translation is intrinsically associated with the notion of path, hence of curve. It might then be natural to consider curves instead of vectors, in order to stay in the considered space when moving from a point to another one. *A priori*, these curves might be geodesics, that is curves of minimal length, in such a way that in Euclidean space, in which geodesics are straight lines, the concept of translation by a vector is kept.

If recourse to geodesics is the evident way for extending the notion of translation, a generalization for other families of curves is even possible, owing to a very powerful mathematical theory, namely that of Lie groups. This theory, which was developed in the 19th century by the Norwegian mathematician Sophus Lie, provides the appropriate tools for describing continuous symmetry of mathematical objects and structures. Employing the technique of differential geometry, its use is manifestly the best way for treating translations on manifolds. In particular it will be shown that a same element of the group of translations, seen as a Lie group, may generate from a point various points in a manifold, according to the chosen parameterization of the latter. As will be shown further, this fact constitutes a capital point in the elaboration of a common theory for displacively modulated and non-modulated structures.

The current model for describing modulated structures, in particular incommensurately modulated ones, is the so-called *superspace model* (Janssen *et al.*, 2004, 2007; Steurer & Haibach, 2001; van Smaalen, 1995, 2007). In such structures, atomic positions of atoms are those of a basic (or average) structure (with a three-dimensional space-group symmetry), to which a periodic wavefunction of the position is applied. In the case where the wavelength of the wavefunction is incommensurate with the cell parameters of the basic structure (*i.e.* the wavelength cannot be written as a rational number times the cell parameters), the structures are called aperiodic as it is no longer possible to describe them as a three-dimensional repetition of a unit cell. The periodicity might, however, be recovered when embedding them into a higher-dimensional space, called *superspace*. In such space, modulated crystals recover a periodicity, in the sense that they can be seen as a repetition of a unit cell in all dimensions. Two main consequences may be evoked:

(*a*) The augmented structure has symmetry, which is called *superspace symmetry*; the symmetry operations may be written in the traditional form: a matrix and a translation part. Note, however, that the matrix part has some restrictions, as the three dimensions in which the crystal exists cannot be mixed with the added (internal) dimensions.

(*b*) The Fourier transform of the augmented structure may be written as the product of the Dirac comb of the reciprocal higher-dimensional lattice and the structure factor of the augmented crystal.

These considerations show that superspace offers the possibility to deal with incommensurate structures (and also quasicrystals) in the same framework as usual three-dimensional periodic functions, the main difference being the number of dimensions.

The symmetry of modulated crystals has not only been described by the superspace formalism. In 1983, J. M. Perez-Mato and co-workers showed that the superspace groups can be introduced through the invariance properties of the so-called *Landau free-energy expansion*, avoiding any reference to the diffraction pattern (Perez-Mato *et al.*, 1984). Twelve years later, Dräger & Mermin (1996) suggested a model within which the symmetry of modulated crystals appears in the *gauge function* expressing the indistinguishability of the Fourier coefficients of the electron density of a structure. If both of these theories do not require a higher-dimensional space for the description of the symmetry of modulated structures, they nevertheless do not offer methods for determining such structures.

In the present work, we show that it is possible to treat the symmetry and obtain the structure factor of displacively modulated structures without adding dimensions. Thanks to Lie theory, incommensurate structures have symmetry in our three-dimensional space. Moreover, as is the case for periodic structures, the three-dimensional Fourier transform of such structures may also be separated into the Fourier transform of one (modulated) unit cell on the one hand and the translational aspect (*i.e.* the translational symmetry) on the other. Thus, modulated structures may be refined without the necessity to work in a higher-dimensional space.

2. The group of translations as a Lie group

In a previous article (Kocian *et al.*, 2009), it was shown that an isometry in the n-dimensional Euclidean manifold [that is in Euclidean space $\mathbb{R}^n$ furnished with the natural coordinate system $u = (u^1; \ldots; u^n)$] acts on any point with natural (Cartesian) coordinates $u = (u^1; \ldots; u^n)$, as

$$u'^i = \sum_{j=1}^{n} f^i{}_j\, u^j + s^i, \tag{1a}$$

where s^i is a constant and $f^i{}_j$ is the component $(i; j)$ of an $n \times n$ constant orthogonal matrix. This relation may be written in a compact matrix notation as

$$u'_p = F u_p + s, \tag{1b}$$

where $s = (s^1; \ldots; s^n) \in \mathbb{R}^n$ is a constant. Equation (1*b*) is often represented in a so-called augmented form, where the matrix and translation parts are combined in one matrix of larger size:

$$\left(\begin{array}{c} u'^1 \\ \vdots \\ \underline{u'^n} \\ 1 \end{array}\right) = \left(\begin{array}{ccc|c} f^1{}_1 & \ldots & f^1{}_n & s^1 \\ \vdots & \ddots & \vdots & \vdots \\ \underline{f^n{}_1} & \ldots & \underline{f^n{}_n} & \underline{s^n} \\ 0 & \ldots & 0 & 1 \end{array}\right) \left(\begin{array}{c} u^1 \\ \vdots \\ \underline{u^n} \\ 1 \end{array}\right).$$

It is very practical but it presents the disadvantage that vectors have an additional component which does not have a physical meaning. An alternative way to combine the matrix and translation part, without adding a dimension, consists in using the derivative technique (Derendinger, 2001). Equation (1*a*) can be written as

$$u'^i = \sum_{j=1}^{n} \left(f^i{}_j + \delta^i{}_j \sum_{l=1}^{n} s^l\, \partial_l \right) u^j, \tag{2a}$$

where $\partial_l \doteqdot \partial/\partial u^l$. In matrix notation

$$u' = \left(F + I_n \sum_{l=1}^{n} s^l\, \partial_l \right) u. \tag{2b}$$

A general isometry in the n-dimensional Euclidean space is then represented by an $n \times n$ matrix $\overline{F} = F + S$, where F is the usual matrix part and S is the $n \times n$ scalar matrix:

$$S = I_n \sum_{l=1}^{n} s^l\, \partial_l. \tag{3}$$

In the case where $F = I_n$, the isometry corresponds to a pure translation, which is represented by the matrix $\overline{S} \doteqdot I_n + S$. The set of all pure translations in the Euclidean manifold is noted $\mathrm{Trans}(\mathbb{R}^n)$. Besides the fact that it is a group, it also has the structure of a smooth manifold (O'Neill, 1983). Indeed, through the trivial parameterization:

$$(s^1; \ldots; s^n) \longmapsto I_n \sum_{l=1}^{n} s^l\, \partial_l,$$

it can be identified to the manifold $\mathbb{R}^n$, as $\{\partial_1; \ldots; \partial_n\}$ forms the canonical basis of vector fields of $\mathbb{R}^n$. The set of all pure translations in $\mathbb{R}^n$ is then, by definition, a *Lie group* which is called a *group of translations* of the Euclidean manifold $\mathbb{R}^n$. According to manifold theory, the set

$$\{W_i \mid 1 \leq i \leq n\},$$

with

$$W_i \doteqdot \left. \frac{\partial \overline{S}}{\partial s^i} \right|_{s=0} = I_n\, \partial_i, \qquad \text{where } s = (s^1; \ldots; s^n), \tag{4}$$

constitutes the canonical basis of the tangent space of the manifold $\mathrm{Trans}(\mathbb{R}^n)$ at the origin ($s = 0$). This space, noted $\mathfrak{trans}(\mathbb{R}^n)$, is not only a vector space but even an algebra. Indeed, besides the addition operation, the vector space is furnished with a bilinear function $\mathfrak{trans}(\mathbb{R}^n) \times \mathfrak{trans}(\mathbb{R}^n) \rightarrow \mathfrak{trans}(\mathbb{R}^n)$, called a *Lie bracket operation*, which has *skew-symmetry* property and which satisfies the *Jacobi identity*. In

this context, the bracket operation is the commutator $[A; B] = AB - BA$, where $A, B \in \mathfrak{trans}(\mathbb{R}^n)$. Thus

$$[W_i; W_j] = 0_n, \quad \forall\, 1 \leq i, j \leq n, \tag{5}$$

as $\partial_i\, \partial_j f = \partial_j\, \partial_i f$, for any function on $\mathbb{R}^n$ the second derivative of which exists.

According to manifold theory, the connection between elements of a Lie group (which is a manifold) and objects of their associated Lie algebra (which is, roughly speaking, the tangent space of the Lie group at the origin) is given by the so-called exponential map. In the case of invertible matrices, the exponential map corresponds to the usual exponential function defined by its power series. Let us try to extend this result to the case of the Euclidean group of translations. Let $V = \sum_{i=1}^{n} s^i\, W_i \in \mathfrak{trans}(\mathbb{R}^n)$, where $s^1, \ldots, s^n$ are the components of V in the canonical basis of $\mathfrak{trans}(\mathbb{R}^n)$; then

$$\exp\left(\sum_{i=1}^{n} s^i\, W_i\right) = \sum_{k=0}^{\infty} \frac{1}{k!} \left(\sum_{i=1}^{n} s^i\, W_i\right)^k,$$

hence

$$\exp\left(\sum_{i=1}^{n} s^i\, W_i\right) = I_n \exp\left(\sum_{i=1}^{n} s^i\, \partial_i\right). \tag{6}$$

This matrix is not equal to $\overline{S} = I_n + S$. However, the expression resulting from its application to the natural coordinates u is equal to $\overline{S}u$, as

$$\begin{aligned}
&\exp\left(\sum_{i=1}^{n} s^i\, \partial_i\right) u^i \\
&= \left[1 + \left(\sum_{i=1}^{n} s^i\, \partial_i\right) + \frac{1}{2}\left(\sum_{i=1}^{n} s^i\, \partial_i\right)^2 + \cdots\right] u^i \\
&= u^i + s^i + 0 + \cdots.
\end{aligned}$$

Thus, the matrix S can be replaced by $\tilde{S} \doteq I_n \exp(\sum_{i=1}^{n} s^i\, \partial_i)$ and $\mathrm{Trans}(\mathbb{R}^n)$ can be defined as

$$\mathrm{Trans}(\mathbb{R}^n) = \left\{ \tilde{S} = I_n \exp\left(\sum_{i=1}^{n} s^i\, \partial_i\right) \;\middle|\; s^i \in \mathbb{R},\ 1 \leq i \leq n \right\}. \tag{7}$$

The interesting point of this formulation is that $\mathrm{Trans}(\mathbb{R}^n)$ does not only generate translations in $\mathbb{R}^n$ but also, at least locally, in any manifold of dimension n embedded in $\mathbb{R}^m$, $m \geq n$. Indeed, let $M \subset \mathbb{R}^m$ be a manifold of dimension $n \leq m$, $\mathcal{U}$ an open set of M, $\tilde{\mathcal{U}}$ an open set of $\mathbb{R}^n$ and

$$\begin{aligned} H : \tilde{\mathcal{U}} &\longrightarrow \mathcal{U} \subset M \\ u &\longmapsto H(u), \end{aligned}$$

$u = (u^1; \ldots; u^n)$ and $H = (h^1; \ldots; h^m)$, a local parameterization of M. Then

$$\begin{aligned}
&\exp\left(\sum_{l=1}^{n} s^l\, \partial_l\right) h^i(u) \\
&= h^i(u) + \sum_{l=1}^{n} s^l\, \frac{\partial h^i(u)}{\partial u^l} \\
&\quad + \frac{1}{2} \sum_{l_1, l_2 = 1}^{n} s^{l_1}\, s^{l_2}\, \frac{\partial^2 h^i(u)}{\partial u^{l_1}\, \partial u^{l_2}} + \cdots \\
&\quad + \frac{1}{r!} \sum_{l_1, \ldots, l_r = 1}^{n} s^{l_1} \cdots s^{l_r}\, \frac{\partial^r h^i(u)}{\partial u^{l_1} \ldots \partial u^{l_r}} + \cdots.
\end{aligned}$$

This expression is exactly the Taylor expansion of h^i in a neighbourhood of a point with natural coordinates u, provided $H(u + s) \in \mathcal{U}$. Thus

$$\exp\left(\sum_{l=1}^{n} s^l\, \partial_l\right) h^i(u) = h^i(u + s), \tag{8}$$

the matrix $\tilde{S}$ carries the point $H(u)$ of M to the point $H(u + s)$ which also belongs to M. For any manifold M of dimension n, embedded in $\mathbb{R}^m$, $n \leq m$, with

$$\begin{aligned} H : \tilde{\mathcal{U}} &\longrightarrow \mathcal{U} \\ u &\longmapsto H(u) \end{aligned}$$

a local parameterization, $\mathcal{U} \subset M$, $\tilde{\mathcal{U}} \subset \mathbb{R}^n$ being two open sets, $u = (u^1; \ldots; u^n)$ and $H = (h^1; \ldots; h^m)$, one can then define a set $\mathrm{Trans}(M)$ as

$$\mathrm{Trans}(M) = \left\{ \tilde{S} = I_m \exp\left(\sum_{l=1}^{n} s^l\, \partial_l\right) \;\middle|\; s^l \in \mathbb{R} \right\}.$$

Any of its elements generates a translation in M provided $H(u + s) \in \mathcal{U}$ when $H(u) \in \mathcal{U}$. $\mathrm{Trans}(M)$ and $\mathrm{Trans}(\mathbb{R}^n)$ are almost the same, the only difference is in the size of the identity matrix. Contrary to $\mathrm{Trans}(\mathbb{R}^n)$, $\mathrm{Trans}(M)$ in general does not have the structure of a group, since certain elements of $\mathrm{Trans}(M)$ carry points of $\mathcal{U} \subset M$ outside $\mathcal{U}$, which is not defined. This problem does not concern us, as we shall henceforth only consider manifolds which can be parameterized by one one-to-one map $H : \mathbb{R}^n \to M$. In such a case, $\mathrm{Trans}(M)$ is a group and is called a *group of translations of* M.

As an example, let us consider the two-dimensional sphere $\mathbb{S}^2 \subset \mathbb{R}^3$ with radius R. Let us take the following parameterization:

$$\begin{aligned}]0; \pi[\times [0; 2\pi[&\longrightarrow \mathbb{S}^2 \setminus \{(0; 0; -R);\ (0; 0; R)\} \\ (\theta;\ \varphi) &\longmapsto (R \sin\theta \cos\varphi;\ R \sin\theta \sin\varphi;\ R \cos\theta) \end{aligned}$$

which covers all points without the north and south poles (in order that H be one-to-one). This sphere $\mathbb{S}^2$ is a typical example of a (two-dimensional) manifold embedded in the three-dimensional Euclidean space. Note that it cannot entirely be parameterized by one one-to-one map. Thus, translations do not form a group in this case. The example is, however, interesting because it concretely shows that translations do not necessarily follow straight lines; they adopt rather the geometry of the considered space. Any element

generating a translation can be written, with respect to the chosen parameterization, as

$$\tilde{S} = I_3 \exp(s^\theta\, \partial_\theta + s^\varphi\, \partial_\varphi),$$

where $\partial_\theta = \partial/\partial\theta$ and $\partial_\varphi = \partial/\partial\varphi$. In the particular case where $s^\varphi = 0$, we have

$$\tilde{S} = I_3 \exp(s^\theta\, \partial_\theta) = I_3 \sum_{k=0}^{\infty} \frac{(s^\theta\, \partial_\theta)^k}{k!}.$$

Applying this matrix to points of $\mathbb{S}^2$, for instance the point with coordinates $(R\sin\theta_1; 0; R\cos\theta_1)$, where $0 < \theta_1 < \pi$, we obtain

$$\begin{aligned}
&\begin{pmatrix} \exp(s^\theta\, \partial_\theta) & 0 & 0 \\ 0 & \exp(s^\theta\, \partial_\theta) & 0 \\ 0 & 0 & \exp(s^\theta\, \partial_\theta) \end{pmatrix} \begin{pmatrix} R\sin\theta\cos\varphi \\ R\sin\theta\sin\varphi \\ R\cos\theta \end{pmatrix}_{(\theta_1;\,0)} \\
&= \begin{pmatrix} R\sin\theta_1 \\ 0 \\ R\cos\theta_1 \end{pmatrix} + s^\theta \begin{pmatrix} R\cos\theta_1 \\ 0 \\ -R\sin\theta_1 \end{pmatrix} \\
&\quad + \frac{(s^\theta)^2}{2!} \begin{pmatrix} -R\sin\theta_1 \\ 0 \\ -R\cos\theta_1 \end{pmatrix} + \frac{(s^\theta)^3}{3!} \begin{pmatrix} -R\cos\theta_1 \\ 0 \\ R\sin\theta_1 \end{pmatrix} + \ldots \\
&= \begin{pmatrix} R\sin\theta_1 \\ 0 \\ R\cos\theta_1 \end{pmatrix} + \frac{(s^\theta)^2}{2!} \begin{pmatrix} -R\sin\theta_1 \\ 0 \\ -R\cos\theta_1 \end{pmatrix} + \ldots \\
&\quad + s^\theta \begin{pmatrix} R\cos\theta_1 \\ 0 \\ -R\sin\theta_1 \end{pmatrix} + \frac{(s^\theta)^3}{3!} \begin{pmatrix} -R\cos\theta_1 \\ 0 \\ R\sin\theta_1 \end{pmatrix} + \ldots \\
&= \begin{pmatrix} R\sin\theta_1\, \cos s^\theta \\ 0 \\ R\cos\theta_1\, \cos s^\theta \end{pmatrix} + \begin{pmatrix} R\cos\theta_1\, \sin s^\theta \\ 0 \\ -R\sin\theta_1\, \sin s^\theta \end{pmatrix}.
\end{aligned}$$

With some trigonometric addition formulae, we finally obtain

$$\begin{aligned}
&\begin{pmatrix} \exp(s^\theta\, \partial_\theta) & 0 & 0 \\ 0 & \exp(s^\theta\, \partial_\theta) & 0 \\ 0 & 0 & \exp(s^\theta\, \partial_\theta) \end{pmatrix} \begin{pmatrix} R\sin\theta\cos\varphi \\ R\sin\theta\sin\varphi \\ R\cos\theta \end{pmatrix}_{(\theta_1;\,0)} \\
&= \begin{pmatrix} R\sin(\theta_1 + s^\theta) \\ 0 \\ R\cos(\theta_1 + s^\theta) \end{pmatrix}.
\end{aligned}$$

This last expression corresponds to the coordinates of a point on $\mathbb{S}^2$ which lies on the same meridian as the starting point $(R\sin\theta_1; 0; R\cos\theta_1)$ (see Fig. 1). Thus, the path linking these two points corresponds to the part of this meridian separating them. The distance between them is therefore equal to the length of this part of the meridian:

$$\ell = \int_{\theta_1}^{\theta_1 + s^\theta} R\, \mathrm{d}t = R s^\theta.$$

In order that the second point stays in the domain of the parameterization, s^θ should be such that $\theta_1 + s^\theta \in]0; \pi[$.

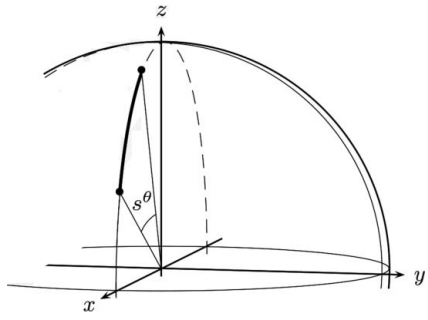


Figure 1
On the sphere, the movement of translation from a point to another one is not along a straight line but along a curve; in this particular case, the curve is a meridian.

2.1. Action of a one-parameter group of translations on a manifold

The action of a group on a vector space is a concept relatively well known, as it is directly connected to *representation theory*. Notwithstanding this, it must be borne in mind that the notion of action of a group is not limited to vector spaces; important applications also exist in the case of manifolds.

For any group $(\mathcal{G}; \diamond)$ with internal composition operation $\diamond$ and neutral element e, a (*left*) *action of* $\mathcal{G}$ *on a manifold* M is a map:

$$\begin{aligned} \mu : \mathcal{G} \times M &\longrightarrow M \\ (g; p) &\longmapsto \mu(g; p) \end{aligned}$$

which satisfies the following conditions:

$$\begin{aligned} \mu(e; p) &= p, && \forall\ p \in M, \\ \mu(g_2; \mu(g_1; p)) &= \mu((g_2 \diamond g_1); p), && \forall\ p \in M, \forall\ g_1, g_2 \in \mathcal{G}. \end{aligned}$$

For the translation group, its action on a manifold M is defined as follows. Let $M \subset \mathbb{R}^m$ be an n-dimensional smooth manifold parameterized by the smooth map $H : \mathbb{R}^n \to M$, $H = (h^1; \ldots; h^m)$. The (left) action μ_T of the group of translation $\mathrm{Trans}(M)$ on M is given by

$$\begin{aligned} \mu_\mathrm{T} : \mathrm{Trans}(M) \times M &\longrightarrow M \\ \big(\tilde{S}; H(u)\big) &\longmapsto \mu_\mathrm{T}\big(\tilde{S}; H(u)\big) = \tilde{S}H(u), \end{aligned}$$

where $H(u)$ is the column vector composed of $h^1(u), \ldots, h^m(u)$ [note that $H(u)$ corresponds to the natural coordinates in $\mathbb{R}^m$ of a point in M]. It is interesting to mention that $\tilde{S}H(u)$ may be written as

$$\tilde{S}H(u) = (H \circ T \circ H^{-1})(H(u)),$$

where T is given by $T(u) = \tilde{S}u$. Indeed, according to the Taylor series, $\tilde{S}H(u) = H(u + s)$, which is exactly what we obtain if we apply $H \circ T \circ H^{-1}$ to $H(u)$, as $\tilde{S}u = u + s$. Rigorously speaking, $\mathrm{Trans}(M)$ is not directly the translation group of the manifold M, but its matrix representation. Elements of this group can also be represented by $H \circ T \circ H^{-1}$, the action of which is defined just above.

In Lie theory, important subgroups of Lie groups are the so-called *one-parameter subgroups*. Such a subgroup in a Lie group $\mathcal{G}$ is a smooth curve c such that $c(t_1 + t_2) = c(t_1)c(t_2)$,

which corresponds to a maximal integral curve of the elements of the associated Lie algebra $\mathfrak{g}$, which start at the neutral element e of the group. The *Lie exponential map* carries elements V of the Lie algebra $\mathfrak{g}$ to $c_V(1) \in \mathcal{G}$ and $t \mapsto \exp(tV)$ is the one-parameter subgroup generated by V (O'Neill, 1983). In the case where the considered Lie group is $\mathrm{GL}_n(\mathbb{R})$ (the set of $n \times n$ invertible matrices), the Lie exponential map is the map exp given by its power series $\exp V = \sum_{k=0}^{\infty} V^k/k!$. Let us extend this result to the group Trans(M) and define one-parameter subgroups as

$$\begin{array}{ccc} \mathbb{R} & \longrightarrow & \mathrm{Trans}(M) \\ t & \longmapsto & \exp(tV), \end{array}$$

where

$$V = \sum_{j=1}^{n} s^j W_j, \quad \text{with } W_j = I_m \, \partial_j.$$

Note that, when $t = 1$, we have $\exp(V)$, that is $\tilde{S}$, the matrix associated with the translation s. The (left) action of such a subgroup of Trans(M) on a point of the manifold M, the coordinates of which are $H(u)$, is

$$(\exp(tV); H(u)) \longmapsto \exp(tV)H(u) = H(u + st),$$

where $s = (s^1; \ldots; s^n)$. Thus, a one-parameter subgroup of translations, that is a curve in Trans(M), generates a curve in M.

2.2. Infinitesimal translation and tangent space

Let $t \mapsto \exp(t\,V)\,H(u) \subset M$ be a curve going through the point $H(u)$, generated by the one-parameter subgroup $t \mapsto \exp(t\,V) \subset \mathrm{Trans}(M)$, $\quad V = \sum_{j=1}^{n} s^j W_j = I_m \sum_{j=1}^{n} s^j \partial_j$, acting on $H(u)$. A point of the curve infinitely close to $H(u)$ is given by

$$\exp(\mathrm{d}t\,V)\,H(u) = H(u + s\,\mathrm{d}t), \tag{9}$$

where $\mathrm{d}t$ is an infinitesimal element of t. Using the Taylor development, we obtain

$$H(u + s\,\mathrm{d}t) \cong H(u) + \sum_{j=1}^{n} \frac{\partial H}{\partial u^j}(u)\, s^j\,\mathrm{d}t \tag{10a}$$

in components:

$$h^i(u + s\,\mathrm{d}t) \cong h^i(u) + \sum_{j=1}^{n} \frac{\partial h^i}{\partial u^j}(u)\, s^j\,\mathrm{d}t. \tag{10b}$$

As $\mathrm{d}t$ is very small, the terms of higher order are negligible. These last two expressions can also be obtained by considering the exponential map series and keeping only the zero and first-order terms (the other being negligible):

$$\exp(\mathrm{d}t\,V) \cong I_m + \mathrm{d}t\,V. \tag{11}$$

The point of coordinates $H(u + s\,\mathrm{d}t)$ can be seen as the tip of a tangent vector at the point of coordinates $H(u)$. Indeed, the tangent vector of the curve $t \mapsto H(u + st)$ at the point of coordinates $H(u)$ is

$$v = \frac{\mathrm{d}}{\mathrm{d}t} H(u + st)\Big|_{t=0} = \sum_{j=1}^{n} \frac{\partial H}{\partial u^j}(u)\, s^j; \tag{12a}$$

in components:

$$v^i = \frac{\mathrm{d}}{\mathrm{d}t} h^i(u + st)\Big|_{t=0} = \sum_{j=1}^{n} \frac{\partial h^i}{\partial u^j}(u)\, s^j. \tag{12b}$$

Multiplying these relations by $\mathrm{d}t$, we obtain an infinitesimal tangent vector at the point of coordinates $H(u)$, with the same direction as v:

$$v\,\mathrm{d}t = \sum_{j=1}^{n} \frac{\partial H(u)}{\partial u^j}(u)\, s^j\,\mathrm{d}t; \tag{13a}$$

in components:

$$v^i\,\mathrm{d}t = \sum_{j=1}^{n} \frac{\partial h^i(u)}{\partial u^j}(u)\, s^j\,\mathrm{d}t; \tag{13b}$$

$v\,\mathrm{d}t$ is exactly the second term of the Taylor development [equation (10a)].

Equation (12a), which can be written as

$$v = \sum_{j=1}^{n} s^j\,\partial_j\,H(u) = \sum_{j=1}^{n} s^j\,W_j\,H(u), \tag{14}$$

where $W_j = I_m\,\partial_j$, shows that a vector of the Lie algebra $\mathfrak{trans}(M)$ generates a vector in the tangent space of M at the point of coordinates $H(u)$. In particular, any basis vector $W_i \in \mathfrak{trans}(M)$ generates the vector

$$w_i = W_i\,H(u) = \frac{\partial H}{\partial u^i}(u), \tag{15}$$

which is the ith canonical basis vector of the tangent space of M at the point of coordinates $H(u)$, associated with the parameterization H. As any tangent space to M has the same dimension as the Lie algebra $\mathfrak{trans}(M)$, we conclude that the map that assigns any canonical basis vector $W_i \in \mathfrak{trans}(M)$ to the canonical basis vector w_i of the tangent space of M at the point of coordinates $H(u)$ is linear and one-to-one. Hence, the Lie algebra $\mathfrak{trans}(M)$, seen as a vector space, and any tangent space of M are isomorphic.

2.3. Lattice in a manifold and in its tangent spaces

The derivations done in the previous subsection show that an infinitesimal translation, belonging to the Lie algebra $\mathfrak{trans}(M)$, generates in the manifold M a point very close to the initial point, which can be seen as the tip of a tangent vector of M at the initial point. When the translation is not infinitesimal, this result in general does not hold any more. Indeed, consider the (smooth) curve:

$$\begin{array}{ccc} [0; 1] & \longrightarrow & \mathfrak{trans}(M) \\ t & \longmapsto & tV = t\sum_{j=1}^{n} s^j W_j. \end{array}$$

The Lie exponential map carries this curve to a curve in the translation group

$$[0;1] \longrightarrow \mathrm{Trans}(M)$$
$$t \longmapsto \exp(tV) = \exp\left(t\sum_{j=1}^{n} s^j W_j\right)$$

which generates a curve in the manifold M:

$$[0;1] \longrightarrow M$$
$$t \longmapsto \exp(tV)H(u) = H(u+st).$$

The tangent vector to this curve at the starting point, with coordinates $H(u)$, is

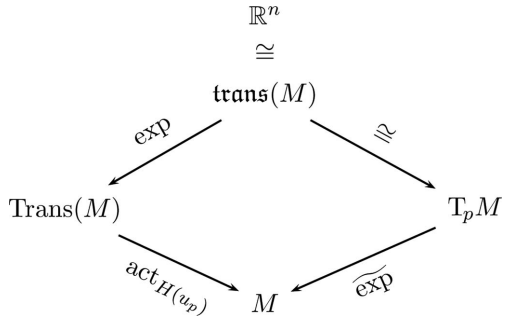
$$v = \sum_{j=1}^{n} \frac{\partial H}{\partial u^j}(u)\, s^j \tag{16}$$

and is not equal to $H(u+s)$ in general. However, even if it is not equal, a one-to-one correspondence between $H(u+s)$ and v can be exhibited. Indeed, since the derivative of a (smooth) map at a point is unique, the tangent vector of a curve at a point is also unique. Reciprocally, let $t \mapsto H(u+rt)$, $r \in \mathbb{R}^n$ be a family of curves which intersect only at $H(u)$; for a given vector v in the tangent space at the point of coordinates $H(u)$, there is, among these curves, exactly one, the tangent vector of which at $t=0$ is v. This vector v can also be obtained when applying the vector $V \in \mathfrak{trans}(M)$ (that is the endpoint of the curve $t \mapsto tV$ parameterized between 0 and 1) to $H(u)$:

$$V\,H(u) = \sum_{j=1}^{n} s^j\, W_j\, H(u) = \sum_{j=1}^{n} s^j\, \frac{\partial H}{\partial u^j}(u) = v. \tag{17}$$

All these considerations can be summarized as follows.

Let $\mathbb{R}^n$ and $\mathbb{R}^m$, $m \geq n$, be, respectively, the n- and m-dimensional Euclidean spaces, and $M \subset \mathbb{R}^m$ a smooth manifold of dimension n, parameterized by the diffeomorphism $H : \mathbb{R}^n \to M$, $H = (h^1; \ldots; h^m)$. Let T_pM be the tangent space of M at the point p of coordinates $H(u_p)$. Let Trans (M) be the translation group of M and $\mathfrak{trans}(M)$ its Lie algebra.
Then



where

$$\exp : \mathfrak{trans}(M) \longrightarrow \mathrm{Trans}(M)$$
$$V \longmapsto \tilde{S} = \exp(V),$$

with $V = \sum_{j=1}^{n} s^j\, W_j = I_m \sum_{j=1}^{n} s^j\, \partial_j$,

$$\mathrm{act}_{H(u_p)}\mathrm{Trans}(M) \longrightarrow M$$
$$\tilde{S} \longmapsto \tilde{S}H(u_p)$$

and

$$\widetilde{\exp}: \quad \mathrm{T}_pM \longrightarrow M$$
$$\left(I_m \sum_{j=1}^{n} s^j\, \partial_j\right)H(u)\Big|_p \longmapsto \exp\left(I_m \sum_{j=1}^{n} s^j\, \partial_j\right)H(u)\Big|_p.$$

$\mathfrak{trans}(M)$ and T_pM are isomorphic through the map

$$\sum_{j=1}^{n} s^j W_j \longmapsto \sum_{j=1}^{n} s^j W_j H(u)\Big|_p, \tag{18a}$$

as well as $\mathbb{R}^n$, seen as a vector space, and $\mathfrak{trans}(M)$, through the map

$$(s^1; \ldots; s^n) \longmapsto \sum_{j=1}^{n} s^j W_j. \tag{18b}$$

It is important to keep in mind the notion of a one-parameter subgroup in Trans(M) and curve in M, even if it does not concretely appear in the previous theorem. The $\widetilde{\exp}$ map carries any vector $v \in \mathrm{T}_pM$ into the point $c_v(1)$ of the curve $t \mapsto c_v(t)$, the expression in coordinates of which is $t \mapsto H(u_p + st)$ and the tangent vector of which is v at $H(u_p)$. As mentioned earlier, there is a one-to-one correspondence between tangent vectors at a point of coordinates $H(u)$ and curves belonging to the family of curves, the expression in coordinates of which is $t \mapsto H(u+rt)$, where $r \in \mathbb{R}^n$. This is due to the fact that there is exactly one straight line linking two points in $\mathbb{R}^n$ and that H is one-to-one. But this does not mean that there is a one-to-one correspondence between tangent vectors v and points $c_v(1)$. This is typically not the case when M is a compact manifold. For instance, on the sphere $\mathbb{S}^2$, an infinity of curves (belonging to the same family, the meridians in this case) link the north to the south poles. Thus, to one point, the south pole, corresponds an infinity of tangent vectors in the tangent space at the north pole, whereas there is only one tangent vector at the north pole which is the tangent vector of a given meridian (linking the north to the south poles).

The previous theorem has an interesting application when a finite free $\mathbb{Z}$-module in $\mathbb{R}^n$ is considered.

Let $M \subset \mathbb{R}^m$ be a manifold parameterized by a diffeomorphism H: $\mathbb{R}^n \to M$, $H = (h^1; \ldots; h^m)$, and T_pM its tangent space at the point p of coordinates $H(u_p)$. Let Trans(M) be the group of translations of M and $\mathfrak{trans}(M)$ its Lie algebra. Let

$$\mathbb{Z}^n_B = \{B\lambda \mid \lambda \in \mathbb{Z}^n\},$$

where $B \in \mathrm{GL}_n(\mathbb{R})$ is an n-dimensional finite free $\mathbb{Z}$-module in $\mathbb{R}^n$. To this module corresponds an n-dimensional finite free $\mathbb{Z}$-module in $\mathfrak{trans}(M)$

$$\mathfrak{Z}^n_B = \left\{\sum_{j=1}^{n} (B\lambda)^j W_j \,\middle|\, \lambda \in \mathbb{Z}^n\right\}, \tag{19}$$

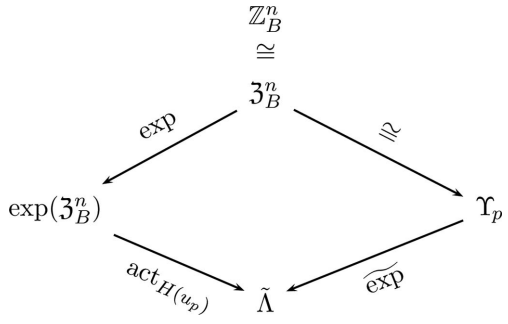
which generates an n-dimensional finite free $\mathbb{Z}$-module

$$\Upsilon_p = \left\{ \sum_{j=1}^{n} \frac{\partial H}{\partial u^j}(u_p)\,(B\,\lambda)^j \;\middle|\; \lambda \in \mathbb{Z}^n \right\}$$

in $\mathrm{T}_p M$ and a discrete subset

$$\tilde{\Lambda} = \{H(u_p + B\lambda) \mid \lambda \in \mathbb{Z}^n\}$$

in M. These results can be summarized in the following diagram.



The discrete subset $\tilde{\Lambda}$ will be called a lattice in the manifold M and the finite free $\mathbb{Z}$-module Υ_p its representation in the tangent space $\mathrm{T}_p M$.

This result is particularly useful for the description of modulated structures. Let us recall that such a structure $\tilde{\mathcal{S}}$ consists of a periodic deformation [through a function (parameterization) H] of a basic (average) structure $\mathcal{S}$ living in the usual n-dimensional Euclidean space. Such a structure may be written as

$$\tilde{\mathcal{S}} = \{H(u_\alpha + B\lambda) \mid 1 \leq \alpha \leq \mu, \lambda \in \mathbb{Z}^n\},$$

where u_α are the coordinates (atomic position) of the atom α in the unit cell of $\mathcal{S}$ and $B \in \mathrm{GL}_n(\mathbb{R})$ is the matrix describing the geometry of the lattice $\{B\lambda \mid \lambda \in \mathbb{Z}^n\}$ associated with $\mathcal{S}$. According to the preceding theorem, $H(u_\alpha + B\lambda)$ can be written as

$$H(u_\alpha + B\lambda) = \exp\left(\sum_{j=1}^{n} (B\lambda)^j W_j\right) H(u)\big|_{u=u_\alpha};$$

hence

$$\tilde{\mathcal{S}} = \left\{ \exp\left(\sum_{j=1}^{n} (B\lambda)^j W_j\right) H(u)\big|_{u=u_\alpha} \;\middle|\; 1 \leq \alpha \leq \mu, \lambda \in \mathbb{Z}^n \right\}.$$

We then arrive at the important conclusion that any structure, be it modulated or not, can be described from one (decorated) unit cell, by applying an *operator* of translation containing a $\mathbb{Z}$-module. This operator is exactly the same for both kinds of structures; the difference appears in its action on points of the unit cell. In the non-modulated case, the map H corresponds to the identity and the action of the operator just consists of adding an element of the $\mathbb{Z}$-module to the coordinates of the point, while in the modulated case, this action also corresponds to a translation, but it takes account of the parameterization H of the space. Thus, thanks to Lie group theory, one has a unified concept of translation for structures, be they modulated or not, without considering any additional dimensions. This claim will become more explicit in the next section.

3. Electron density and Fourier transform

Diffraction by X-rays constitutes one of the most powerful tools for studying crystals at the atomic scale. In a formal mathematical language, diffraction by X-rays corresponds to the calculation of the Fourier transform of the total electron density of all the atoms constituting the crystal.

The appropriate framework for studying the Fourier transform is the Schwartz space $\mathcal{SCH}(\mathbb{R}^n)$ (see Strichartz, 1994), as this space is closed under the Fourier transform operation: the Fourier transform of any function of $\mathcal{SCH}(\mathbb{R}^n)$ is also a function of $\mathcal{SCH}(\mathbb{R}^n)$. However, for crystallographic applications, other functions, which are not in $\mathcal{SCH}(\mathbb{R}^n)$, must be taken into account. Thanks to the theory of distributions, such functions can be treated in the frame of the Schwartz space as well, without creating any problem.

3.1. Structure in the manifold

For an n-dimensional crystal structure, one assumes that the electron density ρ of an atom α is described by a function of $\mathcal{SCH}(\mathbb{R}^n)$, which can be written as

$$\begin{aligned} \mathbb{R}^n &\longrightarrow \mathcal{SCH}(\mathbb{R}^n) \\ r &\longmapsto \rho_\alpha(r - r_\alpha), \end{aligned}$$

where $r = (r^1; \ldots; r^n)$ is an n-dimensional variable and $r_\alpha = (r_\alpha^{\,1}; \ldots; r_\alpha^{\,n})$ are the coordinates of the centre of the considered atom in $\mathbb{R}^n$. These coordinates correspond to the natural coordinates of the n-dimensional Euclidean manifold $\mathbb{R}^n$. The reason for using r here instead of u will become clear further on. The shape of ρ depends on the kind of atom; the function reaches its maximum values in a neighbourhood of the centre of the atom and it vanishes when the norm of $r - r_\alpha$ tends to infinity.

We have seen previously that modulated structures are described in a manifold M which corresponds to $\mathbb{R}^n$, parameterized by the one-to-one map $H : \mathbb{R}^n \to M \cong \mathbb{R}^n$ which is the sum of the identity map and a wavefunction of the position. This function H carries the position u of an atom in the average structure in $\mathbb{R}^n$ to the displaced position $r = H(u) \in \mathbb{R}^n$. A different letter is used for the same natural coordinate system in $\mathbb{R}^n$, in order to distinguish the domain and the target; to be precise, we have $r^j = u^j \circ H(u) = h^j(u)$, where $H = (h^1; \ldots; h^n)$. Electron densities on M are therefore elements of $\mathcal{SCH}(\mathbb{R}^n)$.

Let us consider $r_\alpha = H(u_\alpha)$. Then, the function $r \mapsto \rho_\alpha(r - H(u_\alpha))$ is the electron density of an atom α centred at the point of coordinates $H(u_\alpha)$. It can be written as

$$r \longmapsto \rho_\alpha\big(r - H(u_\alpha + u)\big)\big|_{u=0}. \tag{20}$$

Let $\sum_{j=1}^{n} (B\lambda)^j W_j$ be an element of $\mathfrak{Z}_B^n \subset \mathfrak{trans}(M)$ (see §2.3), where $W_j = I_n\,\partial_j$, with $\partial_j = \partial/\partial u^j$, and B is the matrix char-

acterizing the geometry of the lattice associated with the average structure of the modulated crystal to which the atom α belongs. Then

$$\exp\left(\sum_{j=1}^{n}(B\lambda)^j W_j\right)\rho_\alpha\big(r-H(u)\big)\Big|_{u=u_\alpha}=\rho_\alpha\big(r-H(u_\alpha+B\lambda)\big). \tag{21}$$

Indeed, ρ_α can be considered as a function of $u=(u^1;\ldots;u^n)$:

$$\rho_\alpha\big(r-H(u)\big)=\tilde{\rho}_\alpha(u);$$

thus

$$\begin{aligned}&\exp\left(\sum_{j=1}^{n}(B\,\lambda)^j\,W_j\right)\tilde{\rho}_\alpha(u^1;\ldots;u^n)\\&=\tilde{\rho}_\alpha(u)+\sum_{l=1}^{n}(B\,\lambda)^j\,\frac{\partial\tilde{\rho}_\alpha(u)}{\partial u^j}\\&\quad+\frac{1}{2}\sum_{j_1,j_2=1}^{n}(B\,\lambda)^{j_1}\,(B\,\lambda)^{j_2}\,\frac{\partial^2\tilde{\rho}_\alpha(u)}{\partial u^{j_1}\,\partial u^{j_2}}+\ldots\\&\quad+\frac{1}{r!}\sum_{j_1,\ldots,j_r=1}^{n}(B\,\lambda)^{j_1\cdots}(B\,\lambda)^{j_r}\,\frac{\partial^r\tilde{\rho}_\alpha(u)}{\partial u^{j_1}\ldots\partial u^{j_r}}+\ldots\end{aligned}$$

which exactly corresponds to the Taylor series of $\tilde{\rho}$ about u. Summing equation (21) over $\lambda\in\mathbb{Z}^n$, we obtain the electron density of all the atoms α of the structure. Note that this result may also be obtained by considering $\rho_\alpha(r-H(u_\alpha+u))$ and by taking its value at $u=0$. This point of view is more practical, as the valuation does not depend on α any more; it will therefore be used in the following calculations.

Let us consider the domain

$$\mathcal{D}=\{B\nu\mid\nu=(\nu^1;\ldots;\nu^n),\,0\le\nu^i<1,\,\forall\,1\le i\le n\}.$$

$\mathcal{D}$ is called the *(primitive) unit cell* of the average structure and, by analogy, $\tilde{\mathcal{D}}\doteqdot H(\mathcal{D})$ is the (primitive) unit cell of the corresponding modulated crystal. Suppose that μ atoms (some different, some similar) lie in $\mathcal{D}$ (in the average structure), the centres of which are at the positions u_α, $1\le\alpha\le\mu$. Their positions in the modulated structure are $H(u_\alpha)$. The electron density of all these atoms is then

$$\rho_{\rm cell}(r)=\sum_{\alpha=1}^{\mu}\rho_\alpha\big(r-H(u_\alpha)\big). \tag{22}$$

Let us write

$$\rho_{{\rm cell},u}(r)=\sum_{\alpha=1}^{\mu}\rho_\alpha\big(r-H(u_\alpha+u)\big) \tag{23}$$

and consider $\rho_{\rm cell}$ as a function of u, too. Applying to this latter the exponential of an element $\sum_{j=1}^{n}(B\lambda)^jW_j$, taking its value at $u=0$ and summing over $\lambda\in\mathbb{Z}^n$, we obtain the electron density $\rho(r)$ of the whole modulated structure:

$$\begin{aligned}\rho(r)&=\\&\doteqdot\left[\sum_{\lambda\in\mathbb{Z}^n}\exp\left(\sum_{j=1}^{n}(B\lambda)^jW_j\right)\rho_{{\rm cell},u}(r)\right]_{u=0}\\&=\left[\sum_{\lambda\in\mathbb{Z}^n}\exp\left(\sum_{j=1}^{n}(B\lambda)^jW_j\right)\sum_{\alpha=1}^{\mu}\rho_\alpha\big(r-H(u_\alpha+u)\big)\right]_{u=0}\\&=\left[\sum_{\lambda\in\mathbb{Z}^n}\sum_{\alpha=1}^{\mu}\exp\left(\sum_{j=1}^{n}(B\lambda)^jW_j\right)\rho_\alpha\big(r-H(u_\alpha+u)\big)\right]_{u=0}\\&=\sum_{\lambda\in\mathbb{Z}^n}\sum_{\alpha=1}^{\mu}\rho_\alpha\big(r-H(u_\alpha+B\lambda)\big).\end{aligned} \tag{24}$$

In the case where $H=\mathrm{id}$ (the identity map), we obtain the electron density of the average structure. As in the previous section, we see again that from the electron density of a unit cell, the same set $\mathfrak{Z}_B^n$ generates the electron density of the whole structure, be it modulated or not. Note that this calculation was done under the assumption which consists of taking the electron density of an atom α of the average structure and displacing it in the corresponding modulated structure without turning or distorting it. This is not completely correct, unless the electron densities of the atoms have a spherical symmetry. The conclusion drawn just above is then an approximation of the real case, which is good if the amplitude of the modulation is rather weak (this makes sense, because in the case of a huge amplitude, an average structure corresponding to the modulated one would no longer exist).

In order to find the Fourier transform of ρ, we can either take the last expression of equation (24) and do a traditional calculation, or consider the first expression and exploit the fact that the variable r appears only in $\rho_{{\rm cell},u}$. Following the second way, we obtain

$$\begin{aligned}&(\mathcal{F}\rho)(k)\\&=\int_{\mathbb{R}^n}\left[\sum_{\lambda\in\mathbb{Z}^n}\exp\left(\sum_{j=1}^{n}(B\lambda)^jW_j\right)\rho_{{\rm cell},u}(r)\right]_{u=0}\exp(\mathrm{i}k\cdot r)\,\mathrm{d}^nr\\&=\left[\sum_{\lambda\in\mathbb{Z}^n}\exp\left(\sum_{j=1}^{n}(B\lambda)^jW_j\right)\int_{\mathbb{R}^n}\rho_{{\rm cell},u}(r)\exp(\mathrm{i}k\cdot r)\,\mathrm{d}^nr\right]_{u=0}\\&=\left[\sum_{\lambda\in\mathbb{Z}^n}\exp\left(\sum_{j=1}^{n}(B\lambda)^jW_j\right)\cdot\right.\\&\qquad\left.\cdot\int_{\mathbb{R}^n}\sum_{\alpha=1}^{\mu}\rho_\alpha\big(r-H(u_\alpha+u)\big)\exp(\mathrm{i}k\cdot r)\,\mathrm{d}^nr\right]_{u=0},\end{aligned}$$

where $\cdot$ is the Euclidean scalar product, given by $k\cdot r=\sum_{a=1}^{n}k_ar^a$ (note that $k_a=\sum_{b=1}^{n}\delta_{ab}k^b=k^a$). Up to this point, there was no need to specify H further. In the following calculations, H shall be considered as the sum of the identity and a *displacement* function $\tilde{H}$, that is $H(u)=u+\tilde{H}(u)$. Thus

$$\rho_\alpha\big(r-H(u_\alpha+u)\big)=\rho_\alpha\big(r-u_\alpha-u-\tilde{H}(u_\alpha+u)\big).$$

Considering the change of variable $\tilde{r}=r-u-\tilde{H}(u_\alpha+u)$, we obtain

$$
\begin{aligned}
&(\mathcal{F}\rho)(k)\\
&= \left[\sum_{\lambda\in\mathbb{Z}^n} \exp\left(\sum_{j=1}^{n}(B\lambda)^j W_j\right)\cdot\right.\\
&\quad\left.\cdot \int\limits_{\mathbb{R}^n} \sum_{\alpha=1}^{\mu} \rho_\alpha(\tilde{\boldsymbol{r}}-u_\alpha)\exp\Big(\mathrm{i}k\cdot\big(\tilde{\boldsymbol{r}}+u+\tilde{H}(u_\alpha+u)\big)\Big)\,\mathrm{d}^n\tilde{\boldsymbol{r}}\right]_{u=0}\\
&= \left[\sum_{\lambda\in\mathbb{Z}^n}\sum_{\alpha=1}^{\mu} \exp\left(\sum_{j=1}^{n}(B\lambda)^j W_j\right)\exp\Big(\mathrm{i}k\cdot\big(u+\tilde{H}(u_\alpha+u)\big)\Big)\cdot\right.\\
&\quad\left.\cdot \int\limits_{\mathbb{R}^n} \rho_\alpha(\tilde{\boldsymbol{r}}-u_\alpha)\exp(\mathrm{i}k\cdot\tilde{\boldsymbol{r}})\,\mathrm{d}^n\tilde{\boldsymbol{r}}\right]_{u=0}\\
&= \sum_{\lambda\in\mathbb{Z}^n}\sum_{\alpha=1}^{\mu} \exp\Big(\mathrm{i}k\cdot\big(B\lambda+\tilde{H}(u_\alpha+B\lambda)\big)\Big)\cdot\\
&\quad\cdot \int\limits_{\mathbb{R}^n} \rho_\alpha(\tilde{\boldsymbol{r}}-u_\alpha)\exp(\mathrm{i}k\cdot\tilde{\boldsymbol{r}})\,\mathrm{d}^n\tilde{\boldsymbol{r}}\\
&= \sum_{\lambda\in\mathbb{Z}^n}\sum_{\alpha=1}^{\mu} \exp\big(\mathrm{i}k\cdot(B\lambda)\big)\exp\big(\mathrm{i}k\cdot\tilde{H}(u_\alpha+B\lambda)\big)\cdot\\
&\quad\cdot \int\limits_{\mathbb{R}^n} \rho_\alpha(\tilde{\boldsymbol{r}}-u_\alpha)\exp(\mathrm{i}k\cdot\tilde{\boldsymbol{r}})\,\mathrm{d}^n\tilde{\boldsymbol{r}}. \qquad (25)
\end{aligned}
$$

Note that, in this derivation, the calculation of the integral is completely independent of the exponential of elements of $\mathfrak{Z}_B^n$. Let us now suppose that $\tilde{H}$ is a periodic wavefunction of the position; that is

$$u \longmapsto \tilde{H}(\xi\cdot u),$$

with $\tilde{H}(\xi\cdot u+\overline{T})=\tilde{H}(\xi\cdot u)$, for a $\overline{T}\in\mathbb{R}_+^*$; note that this relation means that $\tilde{h}^j(\xi\cdot u+\overline{T})=\tilde{h}^j(\xi\cdot u)$, for each $1\le j\le n$, where $\tilde{H}=(\tilde{h}^1;\ldots;\tilde{h}^n)$. ξ is an element of $\mathbb{R}^n$ called the wavevector of the wavefunction $\tilde{H}$. Let $U\in \mathrm{SO}_n(\mathbb{R})$ be a matrix such that all components of the column vector $\xi'=U\xi$ are equal to zero, except the first one, the value of which is $\xi'^1=\|\xi\|=(\xi\cdot\xi)^{1/2}$. The same matrix U applied to the column vector u defines $u'=Uu$. Thus

$$\xi\cdot u = {}^{\mathrm{t}}\xi\, u = {}^{\mathrm{t}}\xi'\,{}^{\mathrm{t}}U\,U\,u' = {}^{\mathrm{t}}\xi'\,u' = \|\xi\|u'^1,$$

and $\tilde{H}$ can be seen as a function of u'^1. Since it is periodic, there exists a $T\in\mathbb{R}_+^*$ such that

$$\tilde{H}(u'^1+T)=\tilde{H}\big(\|\xi\|(u'^1+T)\big)=\tilde{H}(\|\xi\|u'^1),$$

with $T=\overline{T}/\|\xi\|$ and $u'^1\longmapsto \exp(\mathrm{i}k\cdot\tilde{H}(u'^1))$ is also a periodic function, with the same period T; indeed

$$
\begin{aligned}
\exp\big(\mathrm{i}k\cdot\tilde{H}(u'^1+T)\big) &= \exp\left(\mathrm{i}\sum_{j=1}^{n}k_j\tilde{h}^j(u'^1+T)\right)\\
&= \exp\left(\mathrm{i}\sum_{j=1}^{n}k_j\tilde{h}^j(u'^1)\right)\\
&= \exp\big(\mathrm{i}k\cdot\tilde{H}(u'^1)\big).
\end{aligned}
$$

It can then be developed into a Fourier series:

$$\exp\big(\mathrm{i}k\cdot\tilde{H}(u'^1)\big)=\sum_{m\in\mathbb{Z}} c_m(k;\xi)\exp(\mathrm{i}m\omega u'^1),$$

where $\omega=2\pi/T$ and

$$
\begin{aligned}
c_m(k;\xi) &= \frac{1}{T}\int_t^{t+T}\exp\big(\mathrm{i}k\cdot\tilde{H}(u'^1)\big)\exp(-\mathrm{i}m\omega u'^1)\,\mathrm{d}u'^1\\
&= \frac{1}{T}\int_t^{t+T}\exp\big(\mathrm{i}k\cdot\tilde{H}(u'^1)-\mathrm{i}m\omega u'^1\big)\,\mathrm{d}u'^1.
\end{aligned}
$$

The expression $\omega u'^1$ can be written

$$\omega u'^1=\frac{2\pi}{T}u'^1=\frac{2\pi}{T\xi_1'}\xi_1'u'^1=\frac{2\pi}{T\|\xi'\|}\xi'\cdot u'=\frac{2\pi}{T\|\xi\|}\xi\cdot u.$$

We can consider $T\|\xi\|=1$. Indeed, even if it were not the case, we could define the vector $\overline{\xi}=\xi/T\|\xi\|$ and consider $\tilde{H}$ as a function of $\overline{\xi}\cdot u$. Thus

$$\exp\big(\mathrm{i}k\cdot\tilde{H}(u'^1)\big)=\sum_{m\in\mathbb{Z}} c_m(k;\xi)\exp(2\pi\mathrm{i}m\xi\cdot u).$$

Since $\exp(\mathrm{i}k\cdot\tilde{H}(u'^1))=\exp(\mathrm{i}k\cdot\tilde{H}(\xi_1'u'^1))=\exp(\mathrm{i}k\cdot\tilde{H}(\xi\cdot u))$, then

$$\exp\big(\mathrm{i}k\cdot\tilde{H}(\xi\cdot u)\big)=\sum_{m\in\mathbb{Z}} c_m(k;\xi)\exp(2\pi\mathrm{i}m\xi\cdot u);$$

in particular

$$
\begin{aligned}
&\exp\Big(\mathrm{i}k\cdot\tilde{H}\big(\xi\cdot(u_\alpha+B\lambda)\big)\Big)\\
&\quad=\sum_{m\in\mathbb{Z}} c_m(k;\xi)\exp\big(2\pi\mathrm{i}m\xi\cdot(u_\alpha+B\lambda)\big).
\end{aligned}
$$

The Fourier transform of ρ then becomes

$$
\begin{aligned}
&(\mathcal{F}\rho)(k)\\
&= \sum_{\lambda\in\mathbb{Z}^n}\sum_{\alpha=1}^{\mu}\exp\big(\mathrm{i}k\cdot(B\lambda)\big)\cdot\\
&\quad\cdot\sum_{m\in\mathbb{Z}} c_m(k;\xi)\exp\big(2\pi\mathrm{i}m\xi\cdot(u_\alpha+B\lambda)\big)\cdot\\
&\quad\cdot\int\limits_{\mathbb{R}^n}\rho_\alpha(\tilde{\boldsymbol{r}}-u_\alpha)\exp(\mathrm{i}k\cdot\tilde{\boldsymbol{r}})\,\mathrm{d}^n\tilde{\boldsymbol{r}}\\
&= \sum_{\lambda\in\mathbb{Z}^n}\sum_{\alpha=1}^{\mu}\sum_{m\in\mathbb{Z}} c_m(k;\xi)\exp(2\pi\mathrm{i}m\xi\cdot u_\alpha)\cdot\\
&\quad\cdot\exp\big(\mathrm{i}(k+2\pi m\xi)\cdot(B\lambda)\big)\cdot\\
&\quad\cdot\int\limits_{\mathbb{R}^n}\rho_\alpha(\tilde{\boldsymbol{r}}-u_\alpha)\exp(\mathrm{i}k\cdot\tilde{\boldsymbol{r}})\,\mathrm{d}^n\tilde{\boldsymbol{r}}.
\end{aligned}
$$

The result of the summation over λ finally gives

$$
\begin{aligned}
(\mathcal{F}\rho)(k) = (2\pi)^n\det B^{-1}\sum_{m\in\mathbb{Z}} c_m(k;\xi)\cdot\\
\cdot\sum_{\ell\in\mathbb{Z}^n}\delta\Big(k-2\pi\big({}^{\mathrm{t}}(B^{-1})\ell-m\xi\big)\Big)\cdot\\
\cdot\sum_{\alpha=1}^{\mu}\int\limits_{\mathbb{R}^n}\rho_\alpha(r-u_\alpha)\exp(\mathrm{i}k\cdot r)\,\mathrm{d}^n r\cdot\\
\cdot\exp(2\pi\mathrm{i}m\xi\cdot u_\alpha), \qquad (26)
\end{aligned}
$$

where δ is the n-dimensional *generalized Dirac function*. This formula shows that the Fourier transform of the electron density of a modulated structure is non-zero only when

$$k = 2\pi\big({}^{\mathrm{t}}(B^{-1})\ell - m\xi\big), \quad \ell \in \mathbb{Z}^n, m \in \mathbb{Z}. \tag{27}$$

In the case where $H = \mathrm{id}$ (the identity map), all coefficients c_m are equal to zero, except $c_0 = 1$. Equation (26) becomes

$$(\mathcal{F}\rho)(k) = (2\pi)^n \det B^{-1} \sum_{\ell\in\mathbb{Z}^n} \delta\big(k - 2\pi\,{}^{\mathrm{t}}(B^{-1})\ell\big)\cdot$$
$$\cdot \sum_{\alpha=1}^{\mu} \int_{\mathbb{R}^n} \rho_\alpha(r - u_\alpha) \exp(\mathrm{i}k\cdot r)\,\mathrm{d}^n r; \tag{28}$$

it is non-zero only when

$$k = 2\pi\,{}^{\mathrm{t}}(B^{-1})\ell, \quad \ell \in \mathbb{Z}^n. \tag{29}$$

The set

$$\Lambda^* = \{2\pi\,{}^{\mathrm{t}}(B^{-1})\ell \mid \ell \in \mathbb{Z}^n\}$$

is called the *reciprocal lattice* of $\Lambda = \{B\lambda \mid \lambda \in \mathbb{Z}^n\}$ which is called the *direct lattice*. In order to eliminate the factor 2π appearing in Λ^*, we introduce the variable $\bar{k} = k/2\pi$. Thus, we define the reciprocal lattice of Λ as

$$\overline{\Lambda}^* = \{{}^{\mathrm{t}}(B^{-1})\ell \mid \ell \in \mathbb{Z}^n\}.$$

The lattice Λ, seen in the tangent space at the origin $\mathrm{T}_o\mathbb{R}^n$, has the structure of a $\mathbb{Z}$-module. A natural basis for Λ is $\{Be_1; \ldots; Be_n\}$, where $e_1, \ldots, e_n$ are the column vectors constituting the canonical basis of $\mathbb{R}^n$ (the components of e_i are all equal to zero, except the ith, the value of which is 1); the vectors ${}^{\mathrm{t}}(B^{-1})\,e_1, \ldots, {}^{\mathrm{t}}(B^{-1})\,e_n$ form a basis of $\overline{\Lambda}^*$. Let us write $a_i = B\,e_i$ and $a_j^* = {}^{\mathrm{t}}(B^{-1})\,e_j$, for all $1 \le i, j \le n$; then

$$a_i \cdot a_j^* = {}^{\mathrm{t}}a_i\,a_j^* = {}^{\mathrm{t}}e_i\,{}^{\mathrm{t}}B\,{}^{\mathrm{t}}(B^{-1})\,e_j = {}^{\mathrm{t}}e_i\,e_j = \delta_{ij} \tag{30a}$$

and

$$a_i^* \cdot a_j = {}^{\mathrm{t}}a_i^*\,a_j = {}^{\mathrm{t}}e_i\,B^{-1}\,B\,e_j = {}^{\mathrm{t}}e_i\,e_j = \delta_{ij}. \tag{30b}$$

These relations appear in a clearer way when considering the components of a_i and a_j^*:

$$a_i = \begin{pmatrix} b_i^1 \\ \vdots \\ b_i^n \end{pmatrix} \quad \text{and} \quad a_j^* = \begin{pmatrix} \overline{b}_1^i \\ \vdots \\ \overline{b}_n^i \end{pmatrix},$$

with $B = (b^i{}_j)_{i,j=1}^n$ and $B^{-1} = (\overline{b}^i{}_j)_{i,j=1}^n$. The sets $\{a_1; \ldots; a_n\}$ and $\{a_1^*; \ldots; a_n^*\}$ are both bases of $\mathrm{T}_o\mathbb{R}^n$ and are, respectively, called the *direct basis* and *reciprocal basis*. Equations (30*a*) and (30*b*) are properties of these bases and express, in a way, the duality between the tangent space $\mathrm{T}_o\mathbb{R}^n$ and the cotangent space $\mathrm{T}_o^*\mathbb{R}^n$, that is the space of all differential maps $\mathrm{d}f$ of $f \in C_0^\infty(\mathbb{R}^n)$. This cotangent space is a vector space and its elements are called *one-forms*. If $(x^1; \ldots; x^n)$ is a coordinate system on a neighbourhood $\mathcal{U}$ of a manifold M, the one-forms $\mathrm{d}x^1, \ldots, \mathrm{d}x^n$ form a dual basis of the coordinate vector fields $\partial_1, \ldots, \partial_n$, where $\partial_i = \partial/\partial x^i$, $1 \le i \le n$, as $\mathrm{d}x^i(\partial_j) = \partial x^i/\partial x^j = \delta^i{}_j$ (where $\delta^i{}_j$ is the Kronecker symbol). Thus, in our case, if we write

$$a_i = \sum_{l=1}^n b^l{}_i\,\partial_l \qquad \text{and} \qquad a^{\sharp j} = \sum_{m=1}^n \overline{b}^j{}_m\,\mathrm{d}u^m \tag{31}$$

with $\partial_l = \partial/\partial u^l$, we have

$$a^{\sharp i}(a_j) = \sum_{m=1}^n \overline{b}^j{}_m\,\mathrm{d}u^m \left(\sum_{l=1}^n b^l{}_i\,\partial_l\right)$$
$$= \sum_{l,m=1}^n \overline{b}^j{}_m\,b^l{}_i\,\mathrm{d}u^m(\partial_l)$$
$$= \sum_{l,m=1}^n \overline{b}^j{}_m\,b^l{}_i\,\delta^m{}_l = \sum_{l=1}^n \overline{b}^j{}_l\,b^l{}_i = \delta^i{}_j.$$

In particular, this relation holds when $a_i, 1 \le i \le n$, and $a^{\sharp j}, 1 \le j \le n$, are valued at the origin o. $(a^{\sharp 1}; \ldots; a^{\sharp n})$ is the dual basis of $(a_1; \ldots; a_n)$. The components of $a^{\sharp i}$ are equal to those of $a_i^*, 1 \le i \le n$. This is the reason why the reciprocal basis is often referred to as the dual basis. In any case, we should keep in mind that the vectors $a_1^*, \ldots, a_n^*$ valued at o belong to $\mathrm{T}_o\mathbb{R}^n$ and not to $\mathrm{T}_o^*\mathbb{R}^n$; this explains why their index is at the bottom and not at the top. In order to show that, under a basis change, it is transformed in the same way as the dual vectors $a^{\sharp i}, 1 \le i \le n$, the index is also often written at the top, so that we have equivalent notations $a_i^* = a^{*i}, 1 \le i \le n$. All these considerations show that the tangent and cotangent spaces at a point are isomorphic. The fact that a_i^* are elements of a tangent and not cotangent space causes a problem with units. Indeed, any vector a_i has a unit of distance, whereas a_i^* has a unit of inverse distance. This problem disappears completely when the dual basis, instead of the reciprocal one, is considered. In this case, $\Lambda \in \mathrm{T}_o\mathbb{R}^n$, $\overline{\Lambda}^* \in \mathrm{T}_o^*\mathbb{R}^n$, and the Fourier transform should be defined as a map from $\mathrm{T}_o\mathbb{R}^n$ to $\mathrm{T}_o^*\mathbb{R}^n$.

As a conclusion of these few calculations, we can express the following result:

Let ρ be the electron density of a crystal structure

$$\tilde{\mathcal{S}} = \{H(u_\alpha + B\lambda) \mid 1 \le \alpha \le \mu, \lambda \in \mathbb{Z}^n\},$$

where $\Lambda = \{B\lambda \mid \lambda \in \mathbb{Z}^n\}$ is the associated lattice of the corresponding average structure $\mathcal{S}$. Let also $\rho_{\mathrm{cell},u}|_{u=0}$ be the electron density of the atoms in one unit cell of $\tilde{\mathcal{S}}$ [see equation (23)] and $\mathcal{F}\rho_{\mathrm{cell},u}|_{u=0}$ its Fourier transform. Then

$$\rho(r) = \sum_{\lambda\in\mathbb{Z}^n} \exp\left(\sum_{j=1}^n (B\,\lambda)^j\,\partial_j\right) \rho_{\mathrm{cell},u}(r)\big|_{u=0} \tag{32a}$$

and

$$(\mathcal{F}\rho)(k) = \sum_{\lambda\in\mathbb{Z}^n} \exp\left(\sum_{j=1}^n (B\,\lambda)^j\,\partial_j\right) (\mathcal{F}\rho_{\mathrm{cell},u})(k)\big|_{u=0}; \tag{32b}$$

the same module $\mathfrak{Z}_B^n \in \mathfrak{trans}(\mathbb{R}^n)$ [see equation (19)] generates ρ from ρ_{cell} and $\mathcal{F}\rho$ from $\mathcal{F}\rho_{\mathrm{cell}}$ (see Fig. 2).

The important point is that these two expressions hold for any map H which is at least piecewise smooth, and therefore also when H corresponds to the identity map. Common formulae for the electron density and its Fourier transform

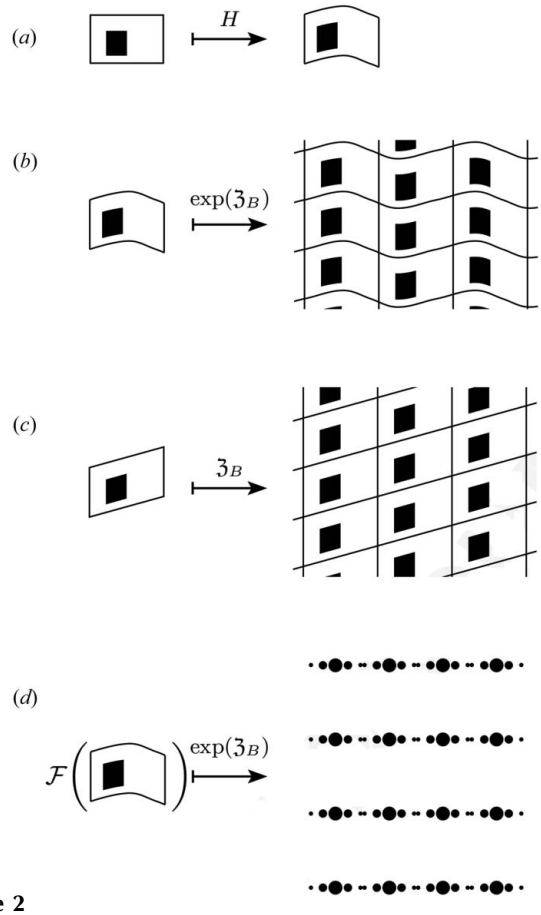


Figure 2
Illustration of the effect of the Lie group of translations. (*a*) Deformation of the average structure through the map H. (*b*) Through the exponential of the module $\mathfrak{Z}_B$, the distorted unit cell generates the whole structure. (*c*) In the tangent space, the unit cell is a parallelepiped and the whole structure is obtained by applying to this cell the module $\mathfrak{Z}_B$. (*d*) The spots appearing in the diffraction pattern are a consequence of the application of the exponential of the module $\mathfrak{Z}_B$ to the Fourier transform of a unit cell.

have been obtained, for non-modulated and (displacive) modulated structures. The electron density of one unit cell, as well as its Fourier transform, appear concretely, with a neat separation with the aspect of translation. Note that, in the non-modulated case, the application of the exponential of $\mathfrak{Z}_B^n$ to the electron density of one unit cell (respectively, its Fourier transform) leads to the convolution (respectively, the normal product) with this electron density (respectively, its Fourier transform) and the Dirac comb of the direct lattice (respectively, the Dirac comb of the reciprocal lattice).

If we have a look at equations (26) and (27), we see that $\mathcal{F}\rho$ is in particular non-zero when $k = 2\pi\,{}^{\mathrm{t}}(B^{-1})\ell$, that is on the node of the reciprocal lattice Λ^* of the lattice Λ associated with the average structure $\mathcal{S}$. Moreover, we notice that the part of equation (26) for which $m = 0$ is identical to equation (28). Thus, the Fourier transform of the electron density of a modulated structure has exactly the same values on the points $k = 2\pi\,{}^{\mathrm{t}}(B^{-1})\ell$ as that of the corresponding average structure. In a way, we can say that the Fourier transform straightens out the shape of a modulated structure; in contrast, it generates additional points. Note that since $\mathcal{F}\rho$ is non-zero when $k = 2\pi\left({}^{\mathrm{t}}(B^{-1})\ell - (m/T\|\xi\|)\xi\right)$, so it is for $|\mathcal{F}\rho|^2$. The points $k = 2\pi\,{}^{\mathrm{t}}(B^{-1})\ell$ are called *main reflections* and all the others for which $\mathcal{F}\rho \neq 0$ are called *satellite reflections*. The index m is called the *order* of the satellite reflection.

3.2. Structure in the tangent space

Let us recall from §2 that the representation $\tilde{\mathcal{T}}_o$ of a modulated structure $\tilde{\mathcal{S}}$ in the tangent space at the origin o is

$$\tilde{\mathcal{T}}_o = \{\Omega(0)(u_\alpha + B\lambda) \mid 1 \leq \alpha \leq \mu, \lambda \in \mathbb{Z}^n\},$$

where $\Omega = (\partial h^i/\partial u^j)_{i,j=1}^n$, $H = (h^1; \ldots; h^n)$. The corresponding electron density can be written as

$${}_{\mathrm{tg}}\rho(v) = \sum_{\lambda\in\mathbb{Z}^n}\sum_{\alpha=1}^{\mu}\rho_\alpha\big(v - \Omega(0)\,(u_\alpha + B\,\lambda)\big). \tag{33}$$

In this expression, the electron density ρ_α of each atom α is considered to be the same as that in the case of the structure in the manifold; it is not turned according to the shape of $\Omega(0)\Lambda$. There is no physical reason for proceeding in such a way, as the structure in the tangent space does not describe reality and is rather a tool for coherent definitions of space- and point-group operations. The use of the same ρ_α is motivated by the wish to have a relatively simple way to connect the electron density of a crystal in the manifold and that in a tangent space (see next subsection).

Geometrically speaking, the representation of a modulated structure in a tangent space has the shape of a non-modulated structure in the n-dimensional Euclidean manifold. This explains why ${}_{\mathrm{tg}}\rho$ can be written as a convolution product [which is in fact a consequence of the application of the module $\mathfrak{Z}_B$ to the electron density of one unit cell in the tangent space ${}_{\mathrm{tg}}\rho_{\mathrm{cell}}$ (see Fig. 2)]:

$${}_{\mathrm{tg}}\rho = {}_{\mathrm{tg}}\rho_{\mathrm{cell}} * \delta_{\Omega(0)\Lambda}, \tag{34}$$

where

$${}_{\mathrm{tg}}\rho_{\mathrm{cell}}(v) = \sum_{\alpha=1}^{\mu}\rho_\alpha\big(v - \Omega(0)u_\alpha\big) \tag{35a}$$

and

$$\delta_{\Omega(0)\Lambda}(v) = \sum_{\lambda\in\mathbb{Z}^n}\delta\big(v - \Omega(0)\lambda\big). \tag{35b}$$

The Fourier transform of ${}_{\mathrm{tg}}\rho$ is then

$$\begin{aligned}(\mathcal{F}_{\mathrm{tg}}\rho)(\kappa) &= (\mathcal{F}_{\mathrm{tg}}\rho_{\mathrm{cell}})(\kappa)(\mathcal{F}\delta_{\Omega(0)\Lambda})(\kappa)\\ &= \int_{\mathbb{R}^n}\sum_{\alpha=1}^{\mu}\rho_\alpha\big(v - \Omega(0)u_\alpha\big)\exp(\mathrm{i}\kappa\cdot v)\,\mathrm{d}^n v\cdot\\ &\quad\cdot\int_{\mathbb{R}^n}\sum_{\lambda\in\mathbb{Z}^n}\delta\big(\tilde{v} - \Omega(0)B\lambda\big)\exp(\mathrm{i}\kappa\cdot\tilde{v})\,\mathrm{d}^n\tilde{v}\\ &= \det\left(B^{-1}\Omega(0)^{-1}\right)\int_{\mathbb{R}^n}\sum_{\alpha=1}^{\mu}\rho_\alpha\big(v - \Omega(0)u_\alpha\big)\exp(\mathrm{i}\kappa\cdot v)\,\mathrm{d}^n v\cdot\\ &\quad\cdot\sum_{\ell\in\mathbb{Z}^n}\delta\Big(\kappa - 2\pi\,{}^{\mathrm{t}}\big(B^{-1}\Omega(0)^{-1}\big)\ell\Big).\end{aligned} \tag{36}$$

This expression is non-zero only when

$$\kappa = 2\pi\,{}^{\mathrm{t}}\big(B^{-1}\Omega(0)^{-1}\big)\ell,\ \ \ell \in \mathbb{Z}^n, \tag{37}$$

that is when

$$\overline{\kappa} = {}^{\mathrm{t}}\big(B^{-1}\Omega(0)^{-1}\big)\ell,\ \ \ell \in \mathbb{Z}^n, \tag{38}$$

where $\overline{\kappa} = \kappa/2\pi$. Note that, in the case where H is the identity map, then

$$(\mathcal{F}_{\mathrm{tg}}\rho)(\kappa) = \det B^{-1} \int\limits_{\mathbb{R}^n} \sum_{\alpha=1}^{\mu} \rho_\alpha(v - u_\alpha)\exp(\mathrm{i}\kappa\cdot v)\,\mathrm{d}^n v\cdot \cdot \sum_{\ell\in\mathbb{Z}^n} \delta\big(\kappa - 2\pi\,{}^{\mathrm{t}}(B^{-1})\ell\big). \tag{39}$$

This equation is identical to equation (28), which is expected, since a crystal structure in the Euclidean manifold and its representation in the tangent space at the origin are exactly the same. In this case, $(\mathcal{F}_{\mathrm{tg}}\rho)$ is non-zero only when

$$\overline{\kappa} = {}^{\mathrm{t}}(B^{-1})\ell,\ \ \ell \in \mathbb{Z}^n. \tag{40}$$

The set

$$\Upsilon_o^* = \{{}^{\mathrm{t}}(B^{-1}\Omega(0)^{-1})\ell \mid \ell \in \mathbb{Z}^n\}$$

is called the *reciprocal lattice* of Υ_o, which is called the *direct lattice*. Both lattices have the structure of a $\mathbb{Z}$-module.

Note that the notions of direct and reciprocal lattices are in fact related to the concepts of direct and reciprocal bases. Natural bases for Υ_o and Υ_o^* are, respectively, $\{\Omega(0)Be_1;\ldots;\Omega(0)Be_n\}$ and $\{{}^{\mathrm{t}}(B^{-1}\Omega(0)^{-1})e_1;\ldots;{}^{\mathrm{t}}(B^{-1}\Omega(0)^{-1})e_n\}$, where $e_1,\ldots,e_n$ are the column vectors forming the canonical basis of $\mathbb{R}^n$. With $\tilde{\boldsymbol{a}}_i = \Omega(0)Be_i$, $1 \le i \le n$, and $\tilde{\boldsymbol{a}}_j^* = {}^{\mathrm{t}}(B^{-1}\Omega(0)^{-1})e_j$, $1 \le j \le n$, we immediately see that $\tilde{\boldsymbol{a}}_i \cdot \tilde{\boldsymbol{a}}_j^* = \tilde{\boldsymbol{a}}_i^* \cdot \tilde{\boldsymbol{a}}_j = \delta_{ij}$. This justifies the names of direct and reciprocal lattices given, respectively, to Υ_o and Υ_o^*. As discussed previously, it is more natural to consider Υ_o as a set in $\mathrm{T}_o M$ and Υ_o^* a set in $\mathrm{T}_o^* M$. Indeed, if we write

$$a_i = \sum_{l=1}^{n} \tilde{b}^{\,l}{}_i\,\partial_l\big|_o \qquad \text{and} \qquad a^{\sharp j} = \sum_{m=1}^{n} \overline{\tilde{b}}^{\,j}{}_m\,\mathrm{d}u^m\big|_o,$$

where $\Omega(0)B = \big(\tilde{b}^{\,i}{}_j\big)_{i,j=1}^n$ and $(\Omega(0)B)^{-1} = \big(\overline{\tilde{b}}^{\,i}{}_j\big)_{i,j=1}^n$, we obtain

$$a^{\sharp i}(a_j) = \sum_{l,m=1}^{n} \overline{\tilde{b}}^{\,j}{}_m\,\tilde{b}^{\,l}{}_i\,\mathrm{d}u^m(\partial_l)\big|_o = \delta^i{}_j.$$

Thus, it appears that the more appropriate framework for the Fourier transform is the Schwartz space on the tangent and cotangent spaces at the origin o of the manifold M:

$$\mathcal{F} : \mathcal{SCH}(\mathrm{T}_o M) \longrightarrow \mathcal{SCH}(\mathrm{T}_o^* M)$$
$$\rho \longmapsto \mathcal{F}\rho.$$

Indeed, $\mathcal{F}$ has a friendly behaviour if the space in which the electron density is considered has the structure of a vector space. In the Euclidean case, any crystal structure in the manifold and its representation in the tangent space at the origin are completely identical; hence the Fourier transform of the electron density in the manifold and in the tangent space at the origin are completely the same. However, for modulated structures, this is more complicated. The previous calculations would suggest considering the electron density of a structure in the tangent space and obtaining its Fourier transform on the cotangent space at the origin; however, this result does not correspond to reality. It would make sense only if a connection between the Fourier transform of the electron density of the real structure and that of its tangent-space representation could be established. This point will be treated in the next subsection. Note that the tangent and cotangent spaces both correspond to $\mathbb{R}^n$, the only difference lying in the base vectors and their units. The canonical basis vectors of $\mathbb{R}^n$ have no unit, while those of $\mathrm{T}_o M$ have a unit of distance and those of $\mathrm{T}_o^* M$ a unit of inverse distance.

3.3. Connection between manifold and tangent space

In equation (33) for ${}_{\mathrm{tg}}\rho$, the argument $\Omega(0)(u_\alpha + B\lambda)$ can be written in components as

$$\sum_{j=1}^{n} \omega^i{}_j(0)\big(u_\alpha{}^j + (B\,\lambda)^j\big) = \sum_{j=1}^{n} \big(u_\alpha{}^j + (B\,\lambda)^j\big)\ \frac{\partial h^i(u)}{\partial u^j}\bigg|_{u=0}, \tag{41}$$

where $\Omega = (\omega^i{}_j)_{i,j=1}^n = (\partial h^i/\partial u^j)_{i,j=1}^n$. To obtain $h^i(u_\alpha + B\,\lambda)$ from this expression, we need to apply the exponential of $\sum_{j=1}^n (u_\alpha^j + (B\,\lambda)^j)\,\partial_j$ to h^i and take the value of the resulting expression at $u = 0$. Since the exponential is given by its power series, it appears, in the connection between ${}_{\mathrm{tg}}\rho$ and ρ, as an infinity of convolution products with Dirac functions. Indeed, let us first consider

$$r \longmapsto \delta\bigg(r - \frac{1}{2}\sum_{j_1,j_2=1}^{n} (u_\alpha + B\,\lambda)^{j_1}\,(u_\alpha + B\,\lambda)^{j_2}\ \frac{\partial^2 H(u)}{\partial u^{j_1}\,\partial u^{j_2}}\bigg). \tag{42}$$

The convolution of this function with

$$r \longmapsto \rho_\alpha\bigg(r - \sum_{j=1}^{n}(u_\alpha + B\,\lambda)^j\,\partial_j H(u)\bigg)$$

is

$$\int\limits_{\mathbb{R}^n} \rho_\alpha\bigg(r - r' - \sum_{j=1}^{n}(u_\alpha + B\,\lambda)^j\ \frac{H(u)}{\partial u^j}\bigg)\cdot \cdot\ \delta\bigg(r' - \frac{1}{2}\sum_{j_1,j_2=1}^{n} (u_\alpha + B\,\lambda)^{j_1}\,(u_\alpha + B\,\lambda)^{j_2}\ \frac{\partial^2 H(u)}{\partial u^{j_1}\,\partial u^{j_2}}\bigg)\,\mathrm{d}^n r',$$

that is

$$\rho_\alpha\bigg(r - \sum_{j=1}^{n}(u_\alpha + B\,\lambda)^j\ \frac{\partial H(u)}{\partial u^j} - \frac{1}{2}\sum_{j_1,j_2=1}^{n} (u_\alpha + B\,\lambda)^{j_1}\,(u_\alpha + B\,\lambda)^{j_2}\ \frac{\partial^2 H(u)}{\partial u^{j_1}\,\partial u^{j_2}}\bigg);$$

the first and second terms of the Taylor series of H appear in the argument of ρ_α. Taking this function and calculating the convolution product with the Dirac function containing the third term of the Taylor series of H, that is

$$\frac{1}{3!}\sum_{j_1,j_2,j_3=1}^{n} (u_\alpha + B\,\lambda)^{j_1}\,(u_\alpha + B\,\lambda)^{j_2}\,(u_\alpha + B\,\lambda)^{j_3}\ \frac{\partial^3 H(u)}{\partial u^{j_1}\,\partial u^{j_2}\partial u^{j_3}},$$

we obtain a function ρ_α in the argument of which the first, second and third terms of the Taylor series appear. Continuing this procedure, we finally obtain a function ρ_α such that all the

terms of the Taylor series of H (the zeroth term, if it is not equal to zero, also needs to be considered) appear in its argument. Let

$$ {}_{\rm tg}\rho_\alpha(r) \doteqdot \rho_{\alpha,u}\left(r - \sum_{j=1}^{n}(u_\alpha + B\,\lambda)^j\,\partial_j H(u)\right) \tag{43}$$

and

$$\delta_{{\rm D}^m_{\alpha,\lambda}H(u)}(r) \doteqdot \delta\left(r - \frac{1}{m!}\sum_{j_1,\ldots,j_m=1}^{n}(u_\alpha + B\,\lambda)^{j_1}\ldots(u_\alpha + B\,\lambda)^{j_m}\,\frac{\partial^m H(u)}{\partial u^{j_1}\ldots\partial u^{j_m}}\right), \tag{44}$$

where the variable v has been replaced by r. Then

$$\rho_\alpha(r) = \left(\delta_{H(u)} * {}_{\rm tg}\rho_{\alpha,u} * \left(\mathop{\ast}_{m=2}^{n}\delta_{{\rm D}^m_{\alpha,\lambda}H(u)}\right)\right)(r)\Big|_{u=0}, \tag{45a}$$

where $\delta_{H(u)}(r) = \delta(r - H(u))$. Note that H can always be considered such that $H(0) = 0$, hence

$$\rho_\alpha(r) = {}_{\rm tg}\rho_{\alpha,u} * \left(\mathop{\ast}_{m=2}^{n}\delta_{{\rm D}^m_{\alpha,\lambda}H(u)}\right)(r)\Big|_{u=0}. \tag{45b}$$

The calculation of the Fourier transform of ρ_α is therefore simply equal to the product of the Fourier transforms of each term of the convolution product:

$$\begin{aligned}
&(\mathcal{F}\rho_\alpha)(k)\\
&= (\mathcal{F}\delta_{H(u)})(k)(\mathcal{F}_{\rm tg}\rho_{\alpha,u})(k)\prod_{m=2}^{n}(\mathcal{F}\delta_{{\rm D}^m_{\alpha,\lambda}H})(k)\Big|_{u=0}\\
&= \exp\left({\rm i}k\cdot H(u)\right)\int_{\mathbb{R}^n}{}_{\rm tg}\rho_{\alpha,u}(r)\exp({\rm i}k\cdot r)\,{\rm d}^n r\cdot\\
&\quad\cdot\prod_{m=2}^{n}\exp\left({\rm i}k\cdot\frac{1}{m!}\sum_{j_1,\ldots,j_m=1}^{n}(u_\alpha + B\lambda)^{j_1}\ldots(u_\alpha + B\lambda)^{j_m}\cdot\right.\\
&\quad\left.\cdot\frac{\partial^m H(u)}{\partial u^{j_1}\ldots\partial u^{j_m}}\right)\Big|_{u=0}.
\end{aligned}$$

This expression almost contains the exponential of the scalar product of ik and the Taylor series of H. To complete it, we multiply $(\mathcal{F}\rho_\alpha)(k)$ by

$$\begin{aligned}
1 &= \exp\left({\rm i}k\cdot\sum_{j=1}^{n}(u_\alpha + B\,\lambda)^j\,\frac{\partial H(u)}{\partial u^j}\right)\cdot\\
&\quad\cdot\exp\left(-{\rm i}k\cdot\sum_{j=1}^{n}(u_\alpha + B\,\lambda)^j\,\frac{\partial H(u)}{\partial u^j}\right)\Big|_{u=0}.
\end{aligned}$$

After simplification, we find

$$\begin{aligned}
(\mathcal{F}\rho_\alpha)(k) &= \int_{\mathbb{R}^n}{}_{\rm tg}\rho_{\alpha,u}(r)\big|_{u=0}\exp({\rm i}k\cdot r)\,{\rm d}^n r\cdot\\
&\quad\cdot\exp\left(-{\rm i}k\cdot\Omega(0)\,(u_\alpha + B\,\lambda)\right)\cdot\\
&\quad\cdot\exp\left({\rm i}k\cdot H(u_\alpha + B\,\lambda)\right).
\end{aligned}$$

Carrying out the change of variable $\tilde{\boldsymbol{r}} - u_\alpha = r - \Omega(0)(u_\alpha + B\lambda)$, we obtain

$${}_{\rm tg}\rho_{\alpha,u}(\tilde{\boldsymbol{r}})\big|_{u=0} = \rho_\alpha(r - u_\alpha),$$

and

$$\begin{aligned}
(\mathcal{F}\rho_\alpha)(k) &= \int_{\mathbb{R}^n}\rho_\alpha(\tilde{\boldsymbol{r}} - u_\alpha)\exp({\rm i}k\cdot\tilde{\boldsymbol{r}})\,{\rm d}^n\tilde{\boldsymbol{r}}\cdot\\
&\quad\cdot\exp(-ik\cdot u_\alpha)\exp\left({\rm i}k\cdot H(u_\alpha + B\lambda)\right)\\
&= \exp\left({\rm i}k\cdot(B\lambda)\right)\exp\left({\rm i}k\cdot\tilde{H}(u_\alpha + B\lambda)\right)\cdot\\
&\quad\cdot\int_{\mathbb{R}^n}\rho_\alpha(\tilde{\boldsymbol{r}} - u_\alpha)\exp({\rm i}k\cdot\tilde{\boldsymbol{r}})\,{\rm d}^n\tilde{\boldsymbol{r}},
\end{aligned}\tag{46}$$

where $H = {\rm id} + \tilde{H}$. Summations over α and λ yield the same relation as equation (25). All these derivations can be summarized as follows.

Let $\tilde{\mathcal{S}}$ be a modulated structure (in the manifold M parameterized by H) and

$$r \longmapsto \rho(r) = \sum_{\lambda\in\mathbb{Z}^n}\sum_{\alpha=1}^{n}\rho_\alpha\big(r - H(u_\alpha + B\lambda)\big).$$

Then

$$\rho(r) = \sum_{\lambda\in\mathbb{Z}^n}\sum_{\alpha=1}^{n}\left(\delta_{H(u)} * {}_{\rm tg}\rho_{\alpha,u} * \left(\mathop{\ast}_{m=2}^{n}\delta_{{\rm D}^m_{\alpha,\lambda}H(u)}\right)\right)(r)\Big|_{u=0}$$

and

$$\begin{aligned}
(\mathcal{F}\rho)(k) &= \sum_{\lambda\in\mathbb{Z}^n}\sum_{\alpha=1}^{n}(\mathcal{F}\delta_{H(u)})(k)(\mathcal{F}_{\rm tg}\rho_{\alpha,u})(k)\cdot\\
&\quad\prod_{m=2}^{n}(\mathcal{F}\delta_{{\rm D}_{\alpha,\lambda^m}H(u)})(k)\Big|_{u=0},
\end{aligned}$$

where ${}_{\rm tg}\rho_{\alpha,u}$ (${}_{\rm tg}\rho_{\alpha,u}|_{u=0}$ being the electron density of the atom α in the tangent space at the origin) and $\delta_{{\rm D}^m_{\alpha,\lambda}H(u)}$ are, respectively, given by equations (43) and (44).

This proposition shows the connection between the electron densities on the manifold and in the tangent space at the origin, and that between the Fourier transforms of the electron densities on the manifold and in the cotangent space at the origin. This result becomes more explicit if we write the expression of the Fourier transform of the electron density in the tangent space at the origin,

$$\begin{aligned}
(\mathcal{F}_{\rm tg}\rho)(k) &= \sum_{\lambda\in\mathbb{Z}^n}\sum_{\alpha=1}^{\mu}\int_{\mathbb{R}^n}\rho_\alpha\big(r - \Omega(0)u_\alpha\big)\exp({\rm i}k\cdot r)\,{\rm d}^n r\cdot\\
&\quad\cdot\exp\left({\rm i}k\cdot(\Omega(0)B\lambda)\right),
\end{aligned}$$

and consider the change of variable $\tilde{\boldsymbol{r}} - u_\alpha = r - \Omega(0)u_\alpha$. Indeed, we obtain

$$\begin{aligned}
(\mathcal{F}_{\rm tg}\rho)(k) &= \sum_{\lambda\in\mathbb{Z}^n}\sum_{\alpha=1}^{\mu}\int_{\mathbb{R}^n}\rho_\alpha\big(r - u_\alpha\big)\exp({\rm i}k\cdot r)\,{\rm d}^n r\cdot\\
&\quad\cdot\exp(-{\rm i}k\cdot u_\alpha)\exp\left({\rm i}k\cdot\big(\Omega(0)(u_\alpha + B\lambda)\big)\right).
\end{aligned}$$

The substitution of $\Omega(0)(u_\alpha + B\lambda)$ by $H(u_\alpha + B\lambda)$ yields the Fourier transform of the electron density of the structure in the manifold. Note that the connection between $\Omega(0)(u_\alpha + B\lambda)$ and $H(u_\alpha + B\lambda)$ is just the Taylor series of H about 0. The summation over the terms of this series appears as a convolution product in the expression for

ρ and as a product in the expression for $\mathcal{F}\rho$, as it must be, since the exponential of a sum is equal to a product of exponentials.

4. Conclusion

The group of translations, seen as a Lie group, is an essential tool for describing the periodicity of a crystal. Any crystal structure, whether modulated or not, is obtained by the application of a discrete subgroup of the translation group to the points of a (decorated) unit cell. The translation group is the instrument that generates the three-dimensional repetition of a brick, while taking into account the particular geometry of the space in which the cell is considered. Thus, the Fourier transform of the electron density of a modulated structure can be written as the Fourier transform of the electron density over one (modulated) unit cell on which acts a discrete subgroup of translations. The notion of *structure factor* is then recovered in modulated structures, without using additional dimensions.

The next step is to try to solve and refine a modulated structure by using the concept of action of the translation group on the electron density over a unit cell. As the structure factor can be isolated in the expression of the Fourier transform of the electron density of a modulated structure, this should be possible somehow, even if the action of the elements of the translation group on a point does not consist of a multiplication but involves the concept of derivative.

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