# Non-Euclidean Symmetries in Crystals

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Crystals also admit a non-Euclidean space-group symmetry. This is shown to be the case for the hexagonal and the cubic-close-packed structures and (slightly broken) for $\alpha$-$Na_2CO_3$. Accordingly, quasicrystals embedded in the superspace and normal crystals in space share the same type of crystallographic symmetries.

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A crystal is a matter distribution with lattice periodicity. That is not the case for quasicrystals. The crystallographic nature of quasicrystals, however, becomes apparent by embedding the structure in a higher $n$-dimensional space, the superspace [1], where it becomes invariant with respect to a lattice of symmetry translations [2].

Indeed, the Fourier wave vectors of a quasicrystal can be expressed as an integral linear combination of $n$ basic ones. The corresponding components form a set of integers, called indices, allowing the labeling of the Bragg diffraction spots. Accordingly, the Fourier components of the quasicrystal structure can be considered as defined by the points of an $n$-dimensional lattice. By an $n$-dimensional Fourier transform, one then gets a periodic structure in one-to-one correspondence with that of the quasicrystal. One has

$$\rho(r) \overset{\mathrm{FT}_3}{\longleftrightarrow} \hat{\rho}(h_1, \ldots, h_n) \equiv \hat{\rho}_s(h_1, \ldots, h_n) \overset{\mathrm{FT}_n}{\longleftrightarrow} \rho_s(r_s) , \qquad (1)$$

where $\mathrm{FT}_n$ denotes the Fourier transform in $n$ dimensions. So quasicrystals (and in general more incommensurate crystals) can be considered as the 3D image of an $n$-dimensional ideal crystal.

The 3D rotational symmetries of a quasicrystal appear in the superspace as $n$-dimensional rotations. In addition, quasicrystals also admit scaling symmetries [3]. Strictly speaking, a quasicrystal is never scaling invariant. In particular, the electronic charge density is a continuous distribution which would be scaling invariant only if constant. So at most a set of discrete points allows a semigroup of scaling-invariant transformations. Here such a set, denoted as a quasicrystal pattern, is defined by the atomic positions of the quasicrystal, the points being labeled by the atomic species. These scaling transformations in space lead to a group of non-Euclidean rotational symmetries in the superspace [4]. Both types of point-group symmetries leave the same lattice invariant. Accordingly, the embedded structure is, in principle, compatible with both Euclidean and non-Euclidean crystallographic transformations.

As normal crystals already have lattice symmetry in three-dimensions, the question then arises whether crystals also admit non-Euclidean symmetries. The aim of the present Letter is to verify with a few concrete examples that this is indeed the case. Considered here are the cubic and the hexagonal close packings. These geometric atomic arrangements are of such general relevance for crystal structures that their non-Euclidean crystallographic symmetries justify the expectation that many more crystals will show these new symmetries.

As follows from general considerations, non-Euclidean crystallographic symmetries, being typically of infinite order, also only apply to crystal structures in the point-atom approximation, with atoms at special positions. This symmetry, therefore, will be very often broken in particular because of the presence of a continuous charge distribution. Nevertheless, these symmetries will impose, if present, relevant structural conditions. How this occurs is shown here by the example of the high-temperature phase $\alpha$-$Na_2CO_3$.

From what has been said, it should be apparent that the aim of this paper is not to introduce new fancy symmetries in crystal physics. Its aim is to find experimental support for the idea that crystallography is characterized by general laws, valid despite the incredible variety in crystal structures, periodic and aperiodic ones.

Several types of three-dimensional non-Euclidean point-group transformations can be defined [5-7]. Here, for simplicity, considerations will be restricted mainly to hyperbolic rotations. These are characterized, like rotations, by an invariant axis (point-wise invariant) and a rotation angle $\chi$. We take a coordinate system spanned by the orthonormal basis vectors $\epsilon_0, \epsilon_1, \epsilon_2$ defining a Minkowskian metric tensor $(g_m)_{\mu\nu} = \epsilon_\mu \epsilon_\nu$, with $\epsilon_0^2 = -1$, $\epsilon_1^2 = \epsilon_2^2 = 1$, and $\epsilon_\mu \epsilon_\nu = 0$ for $\mu \neq \nu$ and choose the invariant axis along $\epsilon_2$. The hyperbolic rotation $A(\chi)$ leaves a lattice invariant if, when expressed with respect to a lattice basis, it is a matrix with integral entries. The crystallographic condition is thus $\mathrm{Tr}\{A(\chi)\} = 1 + 2\cosh\chi = n$ with $n$ integer and $\mathrm{Tr}\{\}$ denoting the trace operation.

*The cubic close packing.*—The cubic close packing consists of atoms at the vertices of a face-centered-cubic lattice $F$ with basis vectors $a_1 = a(0, \frac{1}{2}, \frac{1}{2})$, $a_2 = a(\frac{1}{2}, 0, \frac{1}{2})$, and $a_3 = a(\frac{1}{2}, \frac{1}{2}, 0)$ expressed with respect to a simple cubic lattice with orthonormal basis vectors $e_1, e_2, e_3$. The Euclidean crystallographic symmetry is given by the space group $G = Fm3m$.

For a better understanding of the non-Euclidean symmetry of the lattice $F$ it is convenient to go over first to an orthorhombic body-centered $I$ lattice, spanned by the orthorhombic basis $b_1, b_2, b_3$. The corresponding orientations with respect to the Minkowskian axes of a hyperbolic rotation $A(\chi)$ around $\epsilon_2$ by an angle $\chi$ with $\cosh\chi = 3$ are $b_1 = a(\frac{1}{2}, \frac{1}{2}, 0) = a_3 = (1/\sqrt{2})a\epsilon_2$, $b_2 = a(-\frac{1}{2}, \frac{1}{2}, 0) = a_1 - a_2 = (1/\sqrt{2})a\epsilon_1$, and $b_3 = a(0,0,1) = a_1 + a_2 - a_3 = a\epsilon_0$.

With respect to this basis, $A(\chi)$ becomes the integral matrix $A_b$ leaving invariant the corresponding indefinite metric tensor $g_i(b) = \tilde{A}_b g_i(b) A_b$, with the tilde meaning transposition:

$$A(\chi) \sim A_b = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 3 & 4 \\ 0 & 2 & 3 \end{pmatrix} = \hat{6}\,, \quad g_i(b) = a^2 \begin{pmatrix} \frac{1}{2} & 0 & 0 \\ 0 & \frac{1}{2} & 0 \\ 0 & 0 & -1 \end{pmatrix}. \tag{2}$$

The $I$ lattice is also left invariant, as one sees by considering the body centering $\frac{1}{2}(b_1 + b_2 + b_3) = a(0, \frac{1}{2}, \frac{1}{2})$, or by expressing $A(\chi)$ with respect to the face-centered basis $a_1, a_2, a_3$. Accordingly, the cubic close packing admits non-Euclidean crystallographic symmetries.

The notation $\hat{6}$ adopted in Eq. (2) requires a comment. In three dimensions, hyperbolic rotations (like rotations) are characterized by a point-wise-invariant axis, by the trace, and by the determinant of its matrix representation. A necessary condition for being crystallographic (i.e., for leaving a lattice invariant) is an integral value of the trace, which in the case of rotations is in one-to-one correspondence with the order. It is the order and not the trace which is conventionally used for a characterization, like $4_z$ denoting a rotation by $\pi/2$ around the $z$ axis. Such a convention cannot be used for hyperbolic rotations, because all are of infinite order, even in the crystallographic case. Therefore the trace is used instead. Actually, the two-dimensional trace is adopted, even in three dimensions, because the transformation is essentially a planar one. A caret and an inverted caret allow one to distinguish between proper and improper rotations. So $\hat{n}$ denotes a proper rotation by $2\cos\phi = n$ or by $2\cosh\chi = n$ and $\check{n}$ an improper hyperbolic rotation having $2\sinh\chi = n$. Accordingly, the fourfold rotation $4_z$ is equally well given by $\hat{0}_z$, a sixfold one by $\hat{1}_z$, whereas $\hat{6}_z$ denotes a hyperbolic rotation around the $z$ axis by an angle $\chi = \cosh^{-1}6$.

The symmetry element of infinite order given in Eq. (2) is only one of a fairly large number of other ones. First of all, a conjugation of $A(\chi)$ by the elements of the Euclidean point group $m3m$ leads to additional hyperbolic symmetry rotations. More non-Euclidean symmetries of a cubic lattice can be obtained by considering transformations arising from the self-similarity of a Fibonacci chain and of an octagonal chain, both admitting (as well known) a two-dimensional embedding on a square lattice invariant with respect to hyperbolic rotations [3,4].

To illustrate these additional possibilities, consider a square lattice $\Lambda$ spanned by $a_1 = ae_1$, $a_2 = ae_2$ on an orthonormal basis $e_1, e_2$, and a line $l$ with slope $\tan\alpha = 1/\tau$, where $\tau = (1+\sqrt{5})/2$ is the golden number. Suppose now that $l$ defines an isotropic subspace of the indefinite plane with basis vectors $\epsilon_0, \epsilon_1$ such that $\epsilon_1^2 = -\epsilon_0^2 = 1$ and $\epsilon_0\epsilon_1 = 0$. Then the same lattice $\Lambda$ can be expressed in terms of the new basis according to

$$\begin{aligned} a_1 &= (2/\sqrt{5})^{1/2} a(\cosh\phi\,\epsilon_1 + \sinh\phi\,\epsilon_0)\,, \\ a_2 &= (2/\sqrt{5})^{1/2} a(-\sinh\phi\,\epsilon_1 + \cosh\phi\,\epsilon_0)\,, \end{aligned} \tag{3}$$

with $\tanh\phi = \sqrt{5} - 2$. To the given lattice basis $a_1, a_2$ one can associate two metric tensors, an Euclidean and a non-Euclidean one:

$$g_e = a^2 \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \quad g_i = \frac{2}{\sqrt{5}} a^2 \begin{pmatrix} 1 & -\frac{1}{2} \\ -\frac{1}{2} & -1 \end{pmatrix}. \tag{4}$$

In addition to the fourfold rotation $R$ leaving $g_e$ invariant, one also finds the hyperbolic rotation $A(\chi) = \hat{3}$ with $2\cosh\chi = 3$ leaving $g_i$ invariant:

$$R = \begin{pmatrix} 0 & \bar{1} \\ 1 & 0 \end{pmatrix}, \quad A = \begin{pmatrix} 1 & 1 \\ 1 & 0 \end{pmatrix}^2 = \begin{pmatrix} 2 & 1 \\ 1 & 1 \end{pmatrix} = \hat{3}\,. \tag{5}$$

One has indeed $\tilde{R} g_e R = g_e$ and $\tilde{A} g_i A = g_i$. The connection with the Fibonacci chain follows from the property that the points of the chain can be obtained from the orthogonal projection on the line $l$ of the square-lattice points within a strip parallel to $l$ [8]. The hyperbolic rotation $\hat{3}$, which leaves the square lattice invariant, gives rise on $l$ to a discrete dilatation by $\tau^2$, one of the eigenvalues of $A$. Indeed, $A$ is the square of a matrix expressing the self-similarity of an infinite Fibonacci sequence.

The extension of these considerations to a three-dimensional (simple) cubic lattice is straightforward, and correspondingly also of those applying to other cubic cases.

*Hexagonal close packing.*—The atoms of a hexagonal close packing occupy the $2d$ Wyckoff positions of the space group $G_e = P6_3/mmc$ having as coordinates $\frac{2}{3}, \frac{1}{3}, \frac{1}{4}$ and $\frac{1}{3}, \frac{2}{3}, \frac{3}{4}$ with respect to a hexagonal lattice $P$ with a $c/a$ ratio equal to $\frac{2}{3}\sqrt{6}$. Considering a Minkowskian basis $\epsilon_0, \epsilon_1, \epsilon_2$ oriented as the Euclidean orthonormal ones $e_2, e_1, e_3$, respectively, one can express the hexagonal lattice basis in terms of both of these two coordinate systems:

$$\begin{aligned} a_1 &= a(1,0,0) = a\epsilon_1\,, \\ a_2 &= a(-1/2, \sqrt{3}/2, 0) = a(-(1/2)\epsilon_1 + (\sqrt{3}/2)\epsilon_0)\,, \\ a_3 &= c(0,0,1) = c\epsilon_2\,. \end{aligned}$$

The Euclidean metric tensor and the indefinite one of the hexagonal basis are then

$$g_e = a^2 \begin{pmatrix} 1 & -\frac{1}{2} & 0 \\ -\frac{1}{2} & 1 & 0 \\ 0 & 0 & c^2/a^2 \end{pmatrix}, \qquad (6)$$

$$g_i = a^2 \begin{pmatrix} 1 & -\frac{1}{2} & 0 \\ -\frac{1}{2} & -\frac{1}{2} & 0 \\ 0 & 0 & c^2/a^2 \end{pmatrix}.$$

The sixfold rotation $R$ and the hyperbolic rotation $A$ around the $z$ axis by an angle $\chi$ given by $\cosh\chi = 2$ leave the hexagonal lattice invariant and correspondingly the metric tensors $g_e$ and $g_i$. With respect to the hexagonal basis given above, they are expressed by the matrices

$$R = \begin{pmatrix} 1 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix} = 6_z, \quad A = \begin{pmatrix} 3 & 1 & 0 \\ 2 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} = \hat{4}_z. \qquad (7)$$

The compatibility of both for the given hexagonal close packing becomes apparent if one looks at their action on the coordinates of the two occupied positions $2d$: $R$ as well as $A$ transforms the Wyckoff positions $2d$ into the corresponding $2c$ ones (Fig. 1).

One finds an analogous behavior by considering the following two mirror symmetry transformations (one Euclidean, the other hyperbolic) of the hexagonal lattice:

$$m_d = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad \hat{m}_d = \begin{pmatrix} -1 & 0 & 0 \\ 2 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \qquad (8)$$

Furthermore, both hexagonal metric tensors share the additional two mirror symmetries

$$m = \begin{pmatrix} -1 & 1 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad m_z = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix}. \qquad (9)$$

Therefore, there is a remarkable correspondence between the Euclidean and the non-Euclidean space groups leaving the hexagonal close packing invariant. In addition to the given lattice of translations, the two groups are generated by corresponding elements $g_k^e$ of the Euclidean space group $G_e$ and $g_k^i$ of the non-Euclidean one $G_i$, respectively. Using the Seitz notation one has $g_1^e = \{R|0,0,\frac{1}{2}\} = 6_3$ and $g_1^i = \{A|0,0,\frac{1}{2}\} = \hat{4}_{1/2}$; $g_2^e = \{m_d|0,0,\frac{1}{2}\} = c$ and $g_2^i = \{\hat{m}_d|0,0,\frac{1}{2}\} = \hat{c}$; $g_3^e = \{m|0,0,0\} = m = g_3^i$; and $g_4^e = \{m_z|0,0,0\} = m_z = g_4^i$.

In conclusion, a hexagonal close packing has as symmetry the Euclidean space group $G_e = P6_3/mmc$ and the non-Euclidean one $G_i = P\hat{4}_{1/2}/mm\hat{c}$. Additional hyperbolic symmetry rotations are in principle possible. Indeed, preliminary investigations indicate the possibility to relate the ratio $c/a = \frac{2}{3}\sqrt{6}$ to a non-Euclidean symmetry.

*Hexagonal phase α-Na₂CO₃.*—The high-temperature phase $\alpha$-$Na_2CO_3$ has the same two symmetry space groups as the hexagonal close packing. The non-Euclidean space group, however, does not describe an exact symmetry of the crystal, even in the point atom approximation, because of the expected symmetry-breaking effects.

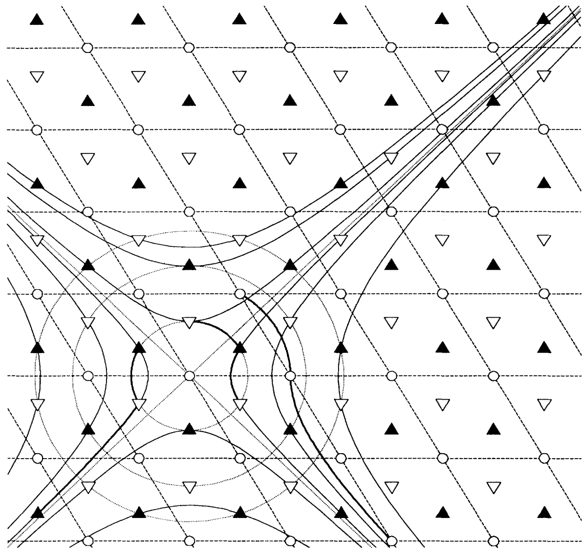

FIG. 1. A rotation by $\pi/3$ and a hyperbolic rotation by $\cosh^{-1}2$, both leave a same hexagonal lattice invariant. These transformations permute the points with fractional coordinates $(\frac{1}{3},\frac{2}{3},z)$ and $(\frac{2}{3},\frac{1}{3},z)$ at the location of the threefold symmetry axes. Accordingly, the product of these two transformations is a point symmetry of the pattern, as indicated by bold lines.

With respect to the space group $G_e = P6_3/mmc$ the following atomic positions are occupied in the $\alpha$ phase [9]:

| | | | | |
|---|---|---|---|---|
| $2a$ | Na(I) | $0,0,0$ | $0,0,\frac{1}{2}$ | |
| $2d$ | Na(II) | $\frac{1}{3},\frac{2}{3},\frac{3}{4}$ | $\frac{2}{3},\frac{1}{3},\frac{1}{4}$ | |
| $2c$ | C | $\frac{2}{3},\frac{1}{3},\frac{3}{4}$ | $\frac{1}{3},\frac{2}{3},\frac{1}{4}$ | |
| $6h$ | O | $x,2x,\frac{1}{4}$ | $2\bar{x},\bar{x},\frac{1}{4}$ | $x,\bar{x},\frac{1}{4}$ |
| | | $\bar{x},2\bar{x},\frac{3}{4}$ | $2x,x,\frac{3}{4}$ | $\bar{x},x,\frac{3}{4}$ |

The corresponding equivalent positions for the non-Euclidean space group $G_i = P\hat{4}_{1/2}/mm\hat{c}$ are the same for $2a$, $2c$, and $2d$, but the sixfold multiplicity of the $6h$ positions requires the special parameter value $x = \frac{1}{6}$. This value is only approximately realized in $\alpha$-$Na_2CO_3$. Experimentally [10] one finds $x = 0.202$ instead of $x = 0.167$, a deviation of about 20%. Considering the essentially different nature of Euclidean and non-Euclidean crystallography and the strong chemical bonds within the $CO_3$ group, this agreement is nevertheless impressive. Furthermore, in the pointlike approximation of this anion, the non-Euclidean space group becomes an exact symmetry.

The natural conceptual frame for dealing with non-Euclidean symmetries in crystals is that of a multimetrical space group [11]. This is nearly self-evident when one tries to characterize the crystallography of quasicrystals, taking properly into account possible scaling symmetries as well [4]. In Fig. 1, multimetrical symmetry transformations appear as a rotation followed by a hyperbolic one, as indicated by bold lines connecting symmetry-related points. Here, that concept has been avoided as much as possible, not because it is not relevant for the examples discussed, but to avoid too many new ideas at the same time.

The spontaneous question arises about the physical meaning of these new geometrical symmetries. Preliminary reflections seem to indicate the relevance of the electrons moving in the fixed frame of pointlike nuclei, both at a classical and at a quantum-mechanical level. Classically, one can think of Kepler orbits of charged (light) particles in the Coulomb field of the nuclei. Quantum mechanically, one possibly has to consider a crystallography defined on a complex space. It is clearly too early to speculate further on all of that.

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