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Stratification of multiples of the elementary cell of a polyethylene line chain

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Abstract. The breakdown of translational symmetry of a polyethylene chain, associated with a Peierls-like phase transition, has been analysed using the general principles of 'action of a group on a set'. All possible phase transitions (dimerization, trimerization, or, in general, p -merization) are classified and the corresponding driving modes are derived.

1. Introduction

Several compounds, such as polymers or organic complexes of the TCNQ salt type, exhibit a periodic ordering only along a single direction. Because of this one-dimensional translational invariance, such compounds are referred to as line (or rod) crystals. Physical properties of the line crystals differ appreciably from those of the classical ones (periodic in three dimensions) for two reasons. (i) A weak coupling between chains, accompanying strong interactions within each chain, yields a large anisotropy. (ii) One-dimensional ordering imposes some specific phenomena, which do not occur in three dimensions: Peierls-Frölich phase transitions (Peierls 1955, Rice and Strässler 1973, Allender *et al* 1974), anomalous infrared activity (Rice *et al* 1977, Rice 1979, Graja *et al* 1981), or high-temperature superconductivity (Keller 1975, Jerome and Schultz 1982). Such phenomena arise as a result of a static deformation of the chain, where two, three or more elementary cells join together forming a new effective cell. This corresponds to dimerization, trimerization, or, in general, p -merization of the chain. Such deformations partially break the translational symmetry of the line crystal.

The aim of this paper is an analysis of some group-theoretic aspects of the breaking of translational symmetry of line polymers taking polyethylene as an example. Breakdown of symmetry, described classically within the thermodynamic Landau theory (Landau 1937, Landau and Lifschitz 1968), can be analysed more transparently by some new methods based on general rules of 'action of a group on a set' (Ascher 1977, Michel and Mozrzymas 1977, Michel 1980, Mozrzymas 1988). Within these methods, the symmetry of the system is made transparent by a stratification of the configuration space, and the possible phase transitions are related to appropriate lattices of epikernels.

The simplest case of such a translation, i.e. a dimerization of the polyethylene chain, is given by us elsewhere (Kuźma *et al* 1989). Here we extend this treatment to a more general case of p -merization (trimerization, tetramerization, etc.).

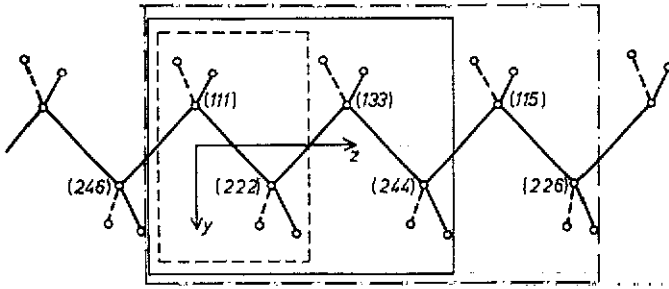


Figure 1. Polyethylene chain; ----, unit; —, double; — · —, threefold elementary cells. Successive numbers in brackets denote carbon atoms in the unit, doubled and threefold elementary cells, respectively

2. The local symmetry group of a p -tuple elementary cell of the polyethylene chain

The polyethylene chain (figure 1) is an example of a line crystal. It can be visualised as an array of carbon and hydrogen atoms, periodic along the invariant axis (the z -axis in figure 1). The full spatial symmetry of an infinite polyethylene chain is given by the line group $L(a) = L2_1/mcm$ in the notation of Vujčić *et al* (1977), a being the lattice constant. Let $T(a) \triangleleft L(a)$ be the maximal translation subgroup of $L(a)$. Then the quotient group

$$Q(a) = L(a)/T(a) \quad (1)$$

describes the geometric distribution of atoms in the one-dimensional elementary cell of the chain.

An important question in the theory of Peierls-like transitions is: how does one extract the order-parameter space from the entire $3N$ n -dimensional configurational space, with N and n being respectively the number of elementary cells and the number of atoms in a single cell. The geometric action of the line group $L(a)$ in the configuration space of the chain decomposes it into subspaces labelled by irreps of $L(a)$. In particular, the subgroup $T(a)$ of $L(a)$ allows us to eliminate translational degrees of freedom, labelled by the quasimomentum which ranges over the Brillouin zone. We are thus left with $3n$ -dimensional secular equations, associated with 'local degrees of freedom' (i.e. Fourier transforms of displacements), corresponding to n atoms in the single elementary cell. These local degrees of freedom are labelled by irreps of the quotient group $Q(a)$.

Such a distinction between local and translational degrees of freedom works perfectly well within the scheme of small crystal vibrations (harmonic phonons), but it is advisable to modify it for the case of Peierls-like phase transitions involving p -merization, i.e. breaking of translational symmetry from $T(a)$ to $T(pa) \triangleleft L(a)$, $p = 2, 3, \dots$. In order to encompass all relevant displacements, one has to introduce a larger quotient group

$$Q(pa) = L(a)/T(pa) \quad (2)$$

which is responsible for the distribution of atoms in the p -merized elementary cell. This group acts in the space of extended local degrees of freedom—the $3pn$ -dimensional space, embracing all possible order parameters, i.e. all modes able to

drive a p -merization. Evidently, the group $Q(pa)$ can be looked at as a finite line group, corresponding to the Born-von Kármán conditions with the period pa . It has the subgroup of 'pure glides', generated by the translation $\varepsilon(a)$ of the period a modulo pa . Pure glides form a cyclic group

$$T_p = \{i\varepsilon(a) \mid i = 1, 2, \dots, p\} \triangleleft Q(pa) \quad (3)$$

with

$$Q(pa)/T_p \cong Q(a). \quad (4)$$

In fact, the group

$$Q(pa) = T_p \square Q(a) \quad (5)$$

is the semidirect product of the point group $Q(a)$ by the group T_p of pure glides. A general element of $Q(pa)$ has the form (α, g) , with $g \in Q(a)$ and

$$\alpha = i\varepsilon(a) + v(g) \quad (6)$$

where $i\varepsilon(a)$ is an integer glide, and $v(g)$ is the fractional translation, associated with the element $g \in Q(a)$, imposed by the structure of the line group $L(a)$ (Sijacki *et al* 1972).

In the case of the polyethylene chain of figure 1 we can establish an isomorphism $\Psi_p : D_{(2p)h} \rightarrow Q(pa)$, $p = 1, 2, \dots$ between the extended point group $Q(pa)$ and the dihedral group $D_{(2p)h}$, defined by

$$\Psi_p(C_{2p}) = (\tfrac{1}{2}, u_z) \quad \Psi_p(\sigma_n) = (0, \sigma_x). \quad (7)$$

In particular, the isomorphisms Ψ_1, Ψ_2 and Ψ_3 , together with the translations α of (6) for $p = 3$ are listed in table 1, with the notation for elements of dihedral groups given in figure 2. This isomorphism can be readily interpreted in terms of Born-von Kármán conditions: the invariant z -axis is 'compactified' to the circumference pa , the z -axis of $Q(pa)$ becomes the principal axis of the dihedral group $D_{(2p)h}$ and the y -axis of $Q(pa)$ corresponds to the radial direction in the cylindrical reference system of the group $D_{(2p)h}$. In this picture, the element in $(\tfrac{1}{2}, u_z)$ generates the cyclic subgroup $C_{2p} \triangleleft D_{(2p)h}$, which, in turn, generates freely the orbit of carbons in the p -tuply enlarged elementary cell. The element $(0, \sigma_x)$ generates the subgroup $C_{1h} \cong \{E, \sigma_n\} \triangleleft D_{(2p)h}$, which yields the hydrogen scissor H_2 for each atom of carbon, so that the subgroup $C_{(2p)h}$ generates freely the orbit of hydrogens. The semidirect product structure (5) becomes now

$$D_{(2p)h} = C_p \square D_{2h} \quad (8)$$

with

$$\Psi_p(C_p) = T_p \quad \Psi_p(D_{2h}) = Q(a) \cong D_{2h} \quad (9)$$

so that, e.g. $\Psi_p(C_{2p}^2) = \varepsilon(a)$, $\Psi_p(C_{2p}^p) = u_x$, $\Psi_p(\sigma_n) = \sigma_x$ etc.

We proceed to perform the stratification of the p -tuple elementary cell along the lines of Kuźma *et al* (1989). The results for $p \leq 3$ are given in tables 2 and 3.

Table 1. Symmetry groups of multiples of elementary cell of polyethylene and their isomorphisms with point groups D_{2h} , D_{4h} and D_{6h} (underlined elements generate the isomorphisms).

$Q(a)$	$p = 1$		$p = 2$		$p = 3$		α
	D_{2h}	$Q(2a)$	D_{4h}	$Q(3a)$	D_{6h}		
E	E	E	E	E	E	0	
		E'	C_2	E'	C_3	1	
				E''	C_3^{-1}	2	
$\underline{\sigma}_x$	$\underline{\sigma}_3(\underline{\sigma}_h)$	$\underline{\sigma}_x$	$\underline{\sigma}_h$	$\underline{\sigma}_x$	$\underline{\sigma}_h$	0	
		σ'_x	I	σ'_x	S_3	1	
				σ''_x	S_3^{-1}	2	
σ_y	I	σ_y	S_4^{-1}	σ_y	S_6	1/2	
		σ'_y	S_4^{-1}	σ'_y	I	3/2	
				σ''_y	S_6^{-1}	5/2	
σ_z	σ_1	σ_z	σ_2	σ_z	σ_5	0	
		σ'_z	σ_4	σ'_z	σ_1	1	
				σ''_z	σ_3	2	
I	σ_2	I	σ_3	I	σ_4	1/2	
		I'	σ_1	I'	σ_6	3/2	
				I''	σ_2	5/2	
\underline{u}_x	\underline{u}_1	\underline{u}_x	\underline{u}_1	\underline{u}_x	\underline{u}_1	1/2	
		u'_x	u_3	u'_x	u_3	3/2	
				u''_x	u_5	5/2	
u_y	u_2	u_y	u_4	u_y	u_2	0	
		u'_y	u_2	u'_y	u_4	1	
				u''_y	u_6	2	
\underline{u}_z	\underline{C}_2	\underline{u}_x	\underline{C}_4	\underline{u}_x	\underline{C}_6	1/2	
		u'_z	\underline{C}_4^{-1}	u'_z	\underline{C}_2	3/2	
				u''_z	c_6^{-1}	5/2	

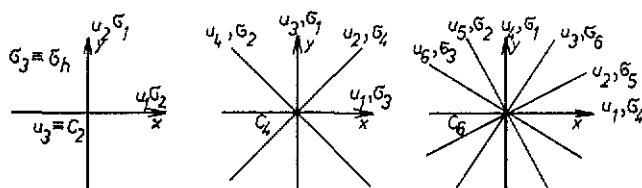


Figure 2. Notation of symmetry elements in D_{2h} , D_{4h} and D_{6h} groups

Each stratum is, by definition, a union of such orbits of the action of the group $Q(pa)$ on the p -merized elementary cell, which are of the same type, labelled by appropriate epikernels, i.e. conjugacy classes of stability groups of points. Thus each stratum corresponds to a kind of 'distinguished points' in the p -merized cell. It results that the stratification does not depend on p . For each p we obtain six strata: the three-dimensional generic stratum $S(C_1)$ (i.e. the stratum with the trivial epikernel C_1), two two-dimensional strata $S(C_2^x)$ and $S(C_2^z)$, two one-dimensional $S(C_2^y)$ and $S(C_{2v}^y)$, and one zero-dimensional $S(C_{2v}^z)$. In particular, the orbit of (the equilibrium positions of) carbon atoms belongs to the stratum $S(C_{2v}^y)$, hydrogen atoms to $S(C_2^z)$, and centres of carbon-carbon bonds (points of inversion symmetry)

Table 2. Stratification of multiples of elementary cells of polyethylene over groups Q(a), Q(2a) and Q(3a).

Stratum and its dimension	Number of elements of the orbit			Parts of a trawers and their dimension	Epikernel				
	p = 1	p = 2	p = 3		p = 1	p = 2	p = 3		
S(C ₁)	3	8	16	24	(0, ∞) × (0, ∞) × (0, 1/4)	3	C ₁	C ₁	C ₁
					(0, ∞) × (0, ∞) × {1/4}	2			
					(0, ∞) × {0} × (0, 1/4)	2			
S(C _s ^x)	2	4	8	12	{0} × (0, ∞) × (0, 1/4)	2	C _s ^x	C _s ^x	C _s ^x
					{0} × (0, ∞) × {1/4}	1			
					{0} × {0} × {0, 1/4}	1			
S(C _s ^z)	2	4	8	12	(0, ∞) × (0, ∞) × {0}	2	C _s ^z	C _s ^z	C _s ^z
					(0, ∞) × {0} × {0}	1			
S(C ₂ ^x)	1	4	8	12	(0, ∞) × {0} × {1/4}	1	C ₂ ^x	C ₂ ^x	C ₂ ^x
S(C _{2v} ^y)	1	2	4	6	{0} × (0, ∞) × {0}	1	C _{2v} ^y	C _{2v} ^y	C _{2v} ^y
S(C _{2v} ^x)	0	2	4	6	{0} × {0} × {0}	0			
					{0} × {0} × {1/4}	0	C _{2v} ^x	C _{2v} ^x	C _{2v} ^x

Table 3. Epikernels of multiples of elementary cell of polyethylene (H - stability subgroup generating an epikernel; for each stratum, the first and the second row of the table gives the elements of the group Q(pa) and the isomorphic point group G, respectively).

Stratum	Q(pa) ≅ G	p = 1 (D _{2h})		p = 2 (D _{4h})		p = 3 (D _{6h})	
		H	ker	H	ker	H	ker
S(C ₁)	Q	{E}	C ₁	{E}	C ₁	{E}	C ₁
	G	{E}	C ₁	{E}	C ₁	{E}	C ₁
S(C _s ^x)	Q	{E, σ _x }	C _s ^x	{E, σ _x }	C _s ^x	{E, σ _x }	C _s ^x
	G	{E, σ ₃ }	C _s ³	{E, σ _h }	C _s ^h	{E, σ _h }	C _s ^h
S(C _s ^z)	Q	{E, σ ₂ }	C _s ^z	{E, σ _x }	C _s ^x	{E, σ _x }	C _s ^x
	G	{E, σ ₁ }	C _s ¹	{E, σ ₂ }	C _s ²	{E, σ ₅ }	C _s ⁵
S(C ₂ ^x)	Q	{E, u _x }	C ₂ ^x	{E, u _x }	C ₂ ^x	{E, u _x }	C ₂ ^x
	G	{E, u ₁ }	C ₂ ¹	{E, u ₁ }	C ₂ ¹	{E, u ₁ }	C ₂ ¹
S(C _{2v} ^y)	Q	{E, u _y , σ _x , σ ₂ }	C _{2v} ^y	{E, u _y , σ _x , σ ₂ }	C _{2v} ^y	{E, u _y , σ _x , σ ₂ }	C _{2v} ^y
	G	{E, u ₂ , σ ₃ , σ ₁ }	C _{2v} ²	{E, u ₂ , σ _h , σ ₂ }	C _{2v} ²	{E, u ₂ , σ _h , σ ₅ }	C _{2v} ⁵
S(C _{2v} ^x)	Q	{E, u _x , I, σ _x }	C _{2v} ^x	{E, u _x , I, σ _x }	C _{2v} ^x	{E, u _x , I, σ _x }	C _{2v} ^x
	G	{E, u ₁ , σ ₂ , σ ₃ }	C _{2v} ¹	{E, u ₁ , σ ₁ , σ _h }	C _{2v} ¹	{E, u ₁ , σ ₄ , σ _h }	C _{2v} ⁴

constitute the zero-dimensional stratum S(C_{2v}^x). It is worth mentioning that the latter stratum can be looked at as a generalization of the invariant point, justifying the name 'point group'. In this context, the group Q(pa) for the polyethylene can be considered as a '2p point group', the factors 2 and p being the result of respectively fractional translations involved in the line group L(a), and of the pure glide group T_p (3). The p-dependence of the stratification consists merely in embedding of an appropriate epikernel in the group Q(pa), and the resulting p-tuplication of the number of elements of each orbit by the group T_p of pure glides.

3. Epikernels and modes driving the polymerization

Let M be the mechanical representation, realising the action of the group $D_{(2p)h}$ in the $3pn$ -dimensional configuration space. Decomposition of M into irreps of $D_{(2p)h}$ (eight one-dimensional $A_{i\alpha}$ and $B_{i\alpha}$, $i = 1, 2$; $\alpha = g$ or u , gerade or ungerade, and $2(p-1)$ two-dimensional $E_{l\alpha}$, $l = 1, 2, \dots, p-1$) yields an appropriate decomposition of the configuration space into irreducible subspaces corresponding to various symmetric modes. The question is which modes are able to drive a p -merization?

Our main result can be formulated as follows: a Peierls-like transition in a polyethylene line chain, consisting in a p -merization of the chain, can be driven only by those irreducible representations E_{lg} or E_{lu} of the group $D_{(2p)h}$ which satisfy the condition

$$\text{LCD}(l, p) = 1 \quad (10)$$

where LCD denotes the largest common divisor. In other words, the only modes active in a p -merization are those belonging to two-dimensional irreps E_{lg} or E_{lu} , l being mutually prime with p .

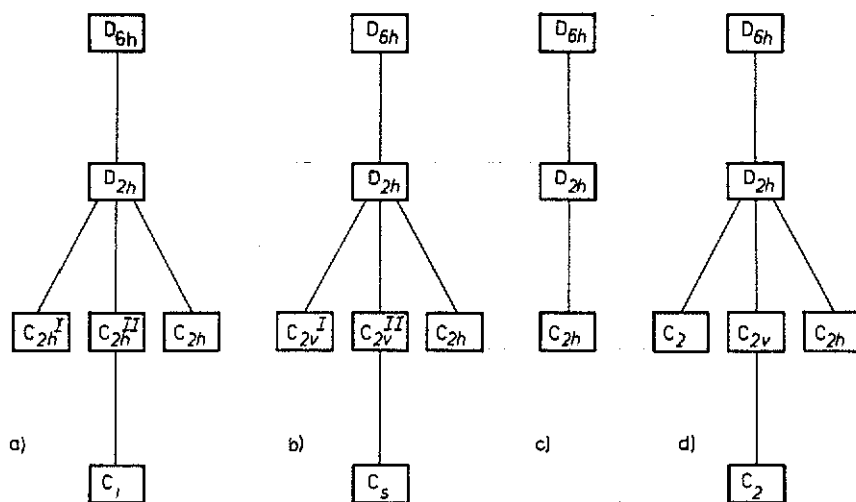


Figure 3. Lattices of epikernels of the D_{6h} point group. (a) epikernel E_{1g} , (b) epikernel E_{1h} (c) epikernel E_{2g} , (d) epikernel E_{2u} .

The proof is based on the observation that the group of broken symmetry cannot enclose—by definition—either the subgroup T_p of pure glides, or any of its non-trivial subgroups $T_\kappa \triangleleft T_p$, $\kappa > 1$ (otherwise one has to have a p' -merization with $p' = p/\kappa$). Thus any broken symmetry can be characterized exclusively by an epikernel given by a subgroup of the quotient group

$$D_{(2p)h}/C_p \cong D_{2h} \quad (11)$$

which eliminates all one-dimensional irreps Γ , since

$$C_p \triangleleft \ker \Gamma \quad \text{for } \dim \Gamma = 1 \quad (12)$$

Table 4. Kernels of irreducible representations of D_{6h} group.

Γ	Ker Γ
A_{1g}	D_{6h}
A_{2g}	$C_{6h} = \{E, 2C_6, 2C_3, C_2, I, 2S_6, 2S_6, \sigma_h\}$
B_{1g}	$D'_{3d} = \{E, 2C_3, 3u', I, 2S_6, 3\sigma'\}$
B_{2g}	$D''_{3d} = \{E, 2C_3, 3u'', I, 2S_6, 3\sigma''\}$
E_{1g}	$C_i = \{E, I\}$
E_{2g}	$C_{2h} = \{E, C_2, I, \sigma_h\}$
A_{1u}	$D_6 = \{E, 2C_6, 2C_3, C_2, 3u', 3u''\}$
A_{2u}	$C_{6v} = \{E, 2C_6, 3C_3, C_2, 3\sigma', 3\sigma''\}$
B_{1u}	$D'_{3h} = \{E, 2C_3, 3u', 2S_3, \sigma_h, 3\sigma''\}$
B_{2u}	$D''_{3h} = \{E, 2C_3, 3u'', 2S_3, \sigma_h, 3\sigma'\}$
E_{1u}	$C_s = \{E, \sigma_h\}$
E_{2u}	$C_2 = \{E, C_2\}$

i.e. each such irrep already contains the group C_p as a subgroup of its kernel. For two-dimensional irreps E_{lg} or E_{lu} , $l = 1, 2, \dots, p - 1$, we have

$$\ker E_{lg} = S_{LCD(l, 2p)} \tag{13}$$

$$\ker E_{lu} = C_{LCD(l, 2p)}. \tag{14}$$

This yields $LCD(l, 2p) = 2$ for epikernels admissible to divide a p -merization, which is equivalent to (10). The case $p = 3$ is presented in figure 3 and table 4.

Table 5. Classification of normal modes of the trimerized elementary cell of polyethylene.

Orbit G			Orbit H					
Λ^C	Δ	Γ^C	Λ^H	Δ	Γ^H	Λ^H	Δ	Γ^H
A_{1g}	A_{2g}	A_{2g}	A_{1g}	A_{2g}	A_{2g}	B_{1u}	A_{2g}	B_{2u}
	B_{2g}	B_{2g}		B_{2g}	B_{2g}		B_{2g}	A_{2u}
	B_{1u}	B_{1u}		B_{1u}	B_{1u}		B_{1u}	A_{1g}
E_{2g}	A_{2g}	E_{2g}	B_{2g}	A_{2g}	B_{1g}	B_{2u}	A_{2g}	B_{1u}
	B_{2g}	E_{1g}		B_{2g}	A_{1g}		B_{2g}	A_{1u}
	B_{1u}	E_{1u}		B_{1u}	A_{2u}		B_{1u}	A_{2g}
B_{1u}	A_{2g}	B_{2u}	E_{1g}	A_{2g}	E_{1g}	E_{1u}	A_{2g}	E_{1u}
	B_{2g}	A_{2u}		B_{2g}	E_{2g}		B_{2g}	E_{2u}
	B_{1u}	E_{2u}					B_{2g}	E_{2g}
E_{1u}	A_{2g}	E_{1u}	E_{2g}	A_{2g}	E_{2g}	E_{2u}	A_{2g}	E_{2u}
	B_{2g}	E_{2u}		B_{2g}	E_{1g}		B_{2g}	E_{1u}
	B_{1u}	E_{2g}		B_{1u}	E_{1u}		B_{1u}	E_{1g}

The driving modes are constructed and classified by the factorization of the mechanical representation (Lulek 1980, Kuźma *et al* 1980, Lulek and Szopa 1990)

$$M = P \otimes V \tag{15}$$

into the positional part

$$P = P_C \oplus P_H \tag{16}$$

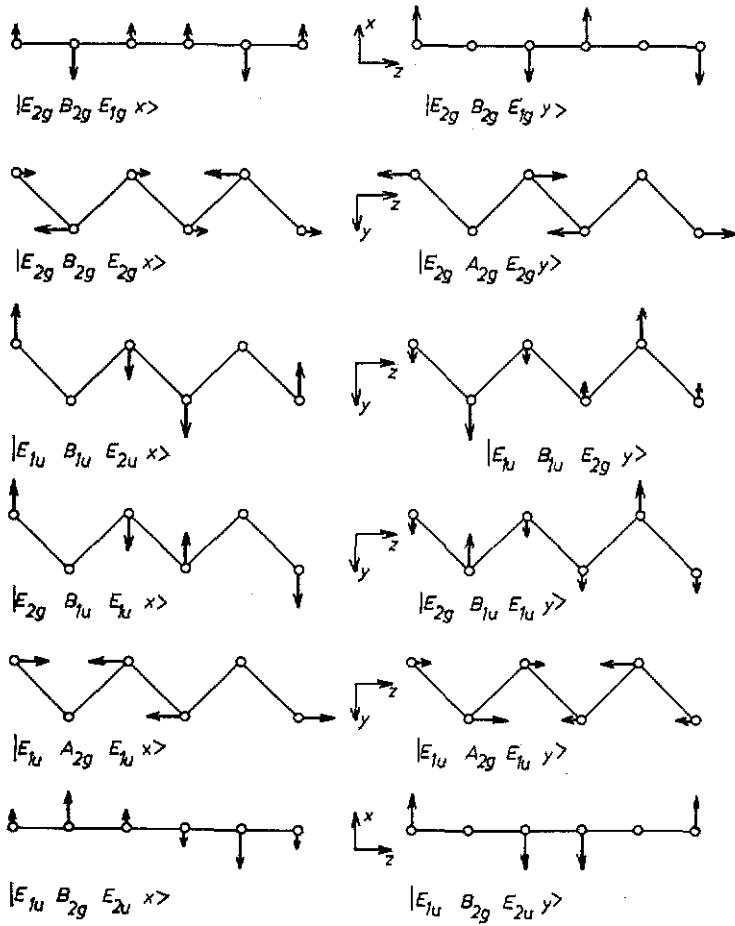


Figure 4. Modes driving the phase transitions in the threefold elementary cell in polyethylene.

consisting of transitive representations

$$P_C = A_{1g} \oplus B_{1g} \oplus \sum_{l=1}^{p-1} E_{lv} \quad v = \begin{cases} g & \text{for } l \text{ even} \\ u & \text{for } l \text{ odd} \end{cases} \quad (17)$$

and

$$P_H = A_{1g} \oplus B_{1g} \oplus A_{2u} \oplus B_{2u} \oplus \sum_{l=1}^{p-1} (E_{lg} \oplus E_{lu}) \quad (18)$$

acting respectively on the orbit of carbons and hydrogens, and the vector factor

$$V = A_{2g} \oplus B_{2g} \oplus B_{1u} \quad (19)$$

acting on the standard displacement fibre (recall that the pure glide group T_p is non-effective in V , so $C_p = \ker V$ —cf Kuzma et al 1989). Thus the modes have the

form

$$|\alpha\Lambda\Gamma\gamma\rangle = \sum_{\lambda\delta} \left[\begin{matrix} \Lambda\Delta\Gamma \\ \lambda\delta\gamma \end{matrix} \right] |\alpha\Lambda\lambda\rangle |\Delta\delta\rangle \quad (20)$$

where $|\alpha\Lambda\lambda\rangle$ and $|\Delta\delta\rangle$ are irreducible bases for the positional and vector representation, respectively, α is the repetition label for Λ in P , and the symbol in square brackets is the Clebsch–Gordan coefficient for the group $D_{(2p)h}$. For $\Gamma = E_{lg}$ or E_{ln} , with l satisfying (10), we obtain from (20) all modes driving the p -merization. The case $p = 3$ is illustrated in table 5 and figure 4.

4. Final remarks

Now we are in a position to provide a detailed answer to the question posed in section 2. The desired subspace of the configuration space of the one-dimensional polyethylene chain, enclosing all modes which drive a Peierls-like phase transition consisting in a p -merization of the elementary cell, is spanned on all those modes given by equation (20), for which the resultant irreps are two-dimensional, $t = E_{lg}$ or E_{ln} , and l is mutually prime with p (equation (10)). Equation (20) provides a systematic way of classifying and constructing a complete orthonormal set of such modes. The order parameter responsible for any p -merization is confined to the space spanned on this set.

A p -merized elementary cell emerges as a result of appropriate modulation of the initial distribution of atoms within the range pa . The symmetry of modulation is related to the quotient group $Q(pa)$ given by (2), with the structure of a finite space group $D_{(2p)h}$, described by (5), (8) and (9). It is the distributing group for the p -merized cell.

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