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2001 J. Phys.: Condens. Matter 13 5399

(http://iopscience.iop.org/0953-8984/13/23/301)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **13** (2001) 5399–5411 www.iop.org/Journals/cm PII: S0953-8984(01)18009-4

Symmetry analysis of static soliton structures and elementary excitations in incommensurately modulated crystals

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Received 13 October 2000, in final form 12 April 2001

Abstract

The most important symmetry properties of the incommensurately modulated crystal structures are investigated by use of exact symmetry theory of quasi-one-dimensional systems in the framework of group theory. It is shown that typical characteristic formulae developed for the description of scattering cross sections of one-dimensionally modulated crystals can be directly derived by the line-group technique. A symmetry analysis of static soliton structures is performed, representing a new method for the investigation of elementary excitations of crystals modulated incommensurately. It leads to the description of symmetry breaking, to the selection rules and hints at the similarity of symmetry behaviour of static and dynamic solitons. The actual formulae for Debye–Waller factors in the case of incommensurately modulated crystals are calculated and tabulated by using generating elements of the line groups concerned.

1. Introduction: aim and scope of line-group theory

Crystals with non-standard structure can also scatter x-rays and neutrons coherently. In an early theory, Dzyaloshinskii (1964, 1965a, b) showed that the classical crystal symmetry description established by Shönfliess, Fedorov and Shubnikov (SFS) cannot reflect the structure of incommensurately modulated crystals. Thus, the rapidly accumulating experimental data on modulated crystals in different areas of solid-state physics (Cummins 1990) led to the elaboration of new mathematical methods for characterization of such crystalline materials based on the superspace method (de Wolff 1974, 1977, de Wolff *et al* 1981), and on the application of cohomology groups (Mermin 1992). Methods of both types have been used successfully to describe some symmetry features of quasicrystals. As has been pointed out for the cases of systems with non-collinear magnetic structure and of commensurate and incommensurate modulations (Kirschner *et al* 1997, 1998), another well-elaborated mathematical method, namely the line-group formalism, significantly extends and improves the efficiency of the investigation of different areas of condensed matter physics. The description of modulated crystals used in the investigation of chaotic behaviour of the anisotropic Ising

0953-8984/01/235399+13$30.00

model with competing interactions, realized by Bak (1981), results in the thermodynamic average for spins M_i at a given site coordinate, i, of a crystal lattice being given by

$$M_i = A\cos\left(\varphi_i + \frac{2\pi \mathrm{i}}{4}\right) \qquad \left(\varphi_i = \frac{\pi}{2}\bar{q}i\right) \tag{1.1}$$

where A denotes the amplitude and the quantity $\bar{q}$ determines the possible values of the discrete phase φ_i describing the modulation. A simple relation between the wave vector $\vec{\bar{q}}$ corresponding to the commensurate phase and $\vec{q}$ relating to the modulation is obtained as

$$q = \frac{\pi}{2}(1+\bar{q})$$

(whenever the vector symbols are not indicated, it is assumed that the relevant vectors are factors in scalar products). This concept has been applied to the examination of incommensurate structures and their chaotic behaviour within the framework of the mean-field approximation (MFA) and with the help of fluctuation theory of phase transitions (Hornreich *et al* 1975, Cowley and Bruce 1978, Bruce and Cowley 1978). According to these studies, the phase transition, which results in modulated crystals, shows critical behaviour characterized by two critical indices, relating to the commensurate and incommensurate phases, respectively. Recent research into the fundamental structural properties of high-temperature superconductors (HTSs) by the neutron diffraction technique (Mook and Doğan 1999, Mook *et al* 2000, Dai *et al* 2000) demonstrates the existence of Q1D subsystems, namely stripes of charge in these materials, which play a significant role in the HTS mechanism (Zaanen 2000). This observation emphasizes the wide applicability of line-group methods, which is confirmed by the results of Damnjanović *et al* (1999a, b) obtained on the symmetry properties of carbon nanotubes.

As has been explained by Hermann (1929) and Vujičić *et al* (1977), the geometrical symmetries of a system, periodic in one direction, always form a line group. Since symmetry transformations of non-crystallographic character are also allowed in this theory (e.g. with screw axes of *rational* order), it is obvious that there are infinitely many line groups, which can be classified into 13 families (Vujičić *et al* 1977). Introduction of the time-reversal symmetry leads to families of coloured symmetry groups of Q1D systems (i.e. the magnetic line groups (Damnjanović and Vujičić 1982)) similarly to in the SFS theory of the ordinary magnetic groups of ideal 3D crystals. For analysis of genuine physical systems, it is necessary to have the irreducible representations (irreps) of line groups, which can be derived by an induction technique applied to Q1D systems (Milošević and Damnjanović 1993, Kirschner *et al* 1997, 1998). Irreps of a full line group L obtained by this procedure and written symbolically as $D^{(\mu)}(L) = D^{(\nu)}(P) \uparrow L$ are obtained from the irreps of self-symmetry point groups P of the motifs and monomers in the case of polymers (unit cells or certain parts of them in the case of crystals). The discrete translations permitted in the line-group theory are *directly coupled* with the orthogonal symmetry transformations of motifs, which is a unique feature of the line-group formalism (Damnjanović and Vujičić 1982).

2. Laying the foundation for this investigation

The aim of our paper is to contribute to the exact theory of Q1D systems by using the line-group method. For this reason, our actual task is to perform symmetry analysis of static soliton structures and elementary excitations, which appear in crystals modulated incommensurately. The basis of this work is provided by our earlier investigations. One of them was devoted to studying the translational symmetry of modulated crystals (Mészáros and Bánkuti 1994). In that paper, the appropriate line groups were determined to characterize the actual symmetry groups belonging to given materials and the structure factors of stereoregular polymers were

extended to modulated systems. Another investigation dealt with the oxygen ordering in high-T_c superconductors (Mészáros *et al* 1997). Four possible superstructures, $L(\bar{4})/2c$, $L(\bar{4})/2m$, $L2/mmm$ and $L2/mcc$, were found for the oxygen symmetry in $Y_1Ba_2Cu_3O_{7-\delta}$ compounds by the line-group method. This provides the possibility of directly comparing the experimental and theoretical diffuse x-ray scattering intensity profiles; an excellent agreement between them is found.

In a later paper we proposed line groups for describing the symmetry properties of systems with non-collinear magnetic structure (Kirschner *et al* 1997). In that work, a general method was elaborated for obtaining the irreps of the symmetry groups of modulated crystals by using those of the line groups. Applying the results obtained on the magnetic superstructure of $MnAu_2$-type compounds, the Dzyaloshinskii invariant was shown to be the most general one for the symmetry properties investigated.

It was also demonstrated that the line-group formalism is suitable for describing both commensurate and incommensurate modulations (Kirschner *et al* 1998). Symmetry groups of modulated crystal lattices were completely characterized by symmetry transformations existing in real space, without application of the formalism based on reciprocal space. As typical examples of the method elaborated, the fundamental invariance and symmetry properties of spin-density functions and soliton lattices were determined in the paper by Kirschner *et al* (1998).

In order to build up a general symmetry theory, that characterizes ideal, modulated and incommensurately modulated crystals in real space (Kirschner *et al* 1998), we have started from a very general formalism of

$$G = S \otimes L \tag{2.1}$$

which means that a space group G can always be created as a direct product of a plane group S and a line group L, where the line group represents the regular arrangements of identical motifs in a given direction.

On the basis of the considerations mentioned above, two demonstration examples are examined, as follows:

(i) *Low-dimensional conductors.* Since the scattering methods play a crucial role in the experimental investigation of low-dimensional conductors (Pouget 1994), these chain-like systems are found among anisotropic materials, where the elementary 'parallel' repeating distance in the chain direction (d_{par}) is smaller than the elementary repeating distance in the perpendicular interchain directions (d_{per}). The 1D metals are unstable at 0 K against a periodic lattice distortion, which results in a modulation of the intrachain electronic density, too (Peierls 1955). The amplitude of the (e.g. x-ray) beam, diffracted by chains possessing elementary structural units that are rigidly displaced along the main axis of the chain by the distance $u = u_{2k_F} \sin(2k_F n d_{par} + \varphi)$, has the form

$$A(\vec{\kappa}) = \sum_n f_n(\vec{\kappa}) \exp\{\mathrm{i}\kappa[n d_{par} + u_{2k_F} \sin(2k_F n d_{par} + \varphi)]\} \tag{2.2}$$

where k_F denotes the wave vector separating occupied electron states from the empty ones and $\vec{\kappa}$ is the elastic scattering vector. It can be rewritten as

$$A(\vec{\kappa}) = \sum_n \sum_\nu f_n(\vec{\kappa}) J_\nu(\kappa u_F) \exp\{\mathrm{i}[(\kappa + 2\nu k_F) n d_{par} + \varphi]\} \tag{2.3}$$

with the help of Bessel-function expansion of its exponential factor. Therefore, the expression for the structure amplitude for low-dimensional conductors is formally identical to the general formula for the structure factor used for description of the elastic scattering of x-rays in the case of stereoregular polymers (Vainshtein 1966), because the periodically

and rigidly repeated structural units of the low-dimensional conductor play the same role as the monomers in the case of such polymers. The formal identity of structure factors is a manifestation of a deep analogy from the point of view of symmetry between systems consisting of bundles of long, straight stereoregular polymer chains and crystals with modulated structure, as is demonstrated in this case.

(ii) *An example from N-dimensional crystallography.* During the structural investigation of the modulated biphenyl crystal (Baudour and Sanquer 1983), the tool of N-dimensional crystallography was applied for the determination and refinement of its structure. It contains long molecular axes, around which an elementary torsion of about 11° appears coupled with a fractional translation of 0.035 Å. Since the coupling of translational and orthogonal symmetry operators is the most general property of line groups, the Seitz operators, which generate the subgroups of generalized translations of line groups, are exactly such symmetry operators, in which the rotations or mirror operations are directly coupled to fractional translations. In this case the structure factor in four-dimensional space can be written in the form

$$F(h_1, h_2, h_3, m) = \sum_n f_n \mathrm{e}^{2\pi \mathrm{i}(h_1 \bar{x}_1^\mu + h_2 \bar{x}_1^\mu + h_3 \bar{x}_3^\mu)} \int_0^1 \mathrm{d}\tau \ \mathrm{e}^{2\pi \mathrm{i} \sum_{j=1}^3 (h_i + m k_i) u_i^\mu(\tau) + m\tau} \tag{2.4}$$

where $\bar{x}_i^\mu$ and u_i^μ denote the average coordinates of undisplaced atoms in the basic unit cell and atomic displacements realizing the modulation, respectively. Taking the particular equation of the modulating atomic displacement field in the form of a sinusoidal function (i.e. let it be $u_i^\mu = U_i^\mu \sin(2\pi\tau - \alpha)$), the integral in (2.4) is transformed into

$$\int_0^1 \mathrm{d}\tau \ \mathrm{e}^{2\pi \mathrm{i} \sum_{j=1}^3 (h_i + m k_i) u_i^\mu(\tau) + m\tau} = \mathrm{e}^{\mathrm{i}m(\alpha+\pi)} J_m \left| 2\pi \sum_{i=1}^3 (h_i + m k_i) U_i^\mu \right| \tag{2.5}$$

where J_m is the Bessel function of order m. The formula (2.4) for the structure amplitude is identical to that introduced for the description of modulated crystal structures by the line-group method (Mészáros and Bánkuti 1994). Within this theory we have proposed and used (Mészáros and Bánkuti 1994, Kirschner *et al* 1998) the following general functional:

$$F_{mod}(\vec{\kappa}) = \mathcal{F}\{f_{mod}(\vec{r}) * \rho(\vec{r})\} = \mathcal{F}\{f_{mod}(\vec{r})\} F(\vec{\kappa}) \tag{2.6}$$

to describe the structure factor of modulated systems, where $*$ denotes the convolution operation, $\rho(\vec{r})$ is the electron-density function of an ideal crystal and $f_{mod}(\vec{r})$ is a line-group-invariant function realizing modulation. This is a functional of very general character and naturally contains the earlier-obtained particular formulae for structure factors of modulated systems. This equivalency can be directly seen from (2.6) and (2.4) rewritten using (2.5) in the form

$$F_l\left(R, \Psi, \frac{l}{c'}\right) = \sum_j f_j \mathrm{e}^{2\pi \mathrm{i} l z_j / c'} \sum_n J_n(2\pi r_j R) \mathrm{e}^{\mathrm{i}[n(\alpha+\pi)]} \qquad \left(\alpha \equiv \Psi - \Psi_j - \frac{\pi}{2}\right). \tag{2.7}$$

In (2.7) the coordinates (R, Ψ, l) and (r_j, Ψ_j, z_j) denote the cylindrical coordinates in reciprocal space and those of the x-ray scattering centres inside the motif in real space, respectively. We use the symbol c' as the elementary translation along the main axis, represented by a line group, instead of c, the elementary translation along the modulation parallel to the crystallographic direction, because these two lengths are generally incommensurate.

3. Investigation of Q1D symmetry properties of soliton structures

In order to demonstrate the line-group invariance of the simplest static soliton structure, we apply the elementary symmetry operator of the subgroup of generalized translations of a line group to the relevant phase function, which represents a new method for the description of different types of modulation. Starting from the MFA of the theory of phase transitions, modulated crystals may appear if the relevant expression for the thermodynamic potential contains derivatives of the order parameter components (in the form of Lifshitz invariants). One such expression is the following power expansion of a thermodynamic potential that corresponds to phase transitions governed by a two-component order parameter:

$$\Phi = \int \mathrm{d}^3\vec{r}\ \left\{ r\rho^2 + u\rho^4 + w\rho^n(1+\cos n\varphi) - \sigma\rho^2\frac{\partial\varphi}{\partial z} + \gamma\left[\left(\frac{\partial\rho}{\partial z}\right)^2 + \rho^2\left(\frac{\partial\varphi}{\partial z}\right)^2\right]\right\} \tag{3.1}$$

given in polar coordinates. Here the coefficients u and w denote respectively the contributions of the isotropic and anisotropic interactions in the system, r is coefficient of the expression containing the sum of the quadratic terms of the order parameter components, whereas σ and γ are determined by the order of successive coefficients of the Lifshitz invariant and sums of terms expressing inhomogeneities in the system being examined. The minimizing of the functional (3.1) with respect to the given phase in the Dzyaloshinskii approximation (Izyumov and Syromyatnikov 1990) leads to the well-known ordinary non-linear differential equation for the mathematical pendulum, having the solution

$$\varphi(z) = \frac{\pi}{n} + \frac{4}{n}\arctan[\mathrm{e}^{\pm n\vec{q}\cdot(\vec{z}-\vec{z}_0)}] \tag{3.2}$$

with z_0 and q constants of integration. (In producing (3.2), a series of refinements of integration constants are performed: the constant appearing at the first integration of the differential equation of the mathematical pendulum is $q^2/2 - v$, by use of which a new integration constant $\kappa^2 = 4v/q^2$ is introduced. For our purposes it is sufficient to examine the limit situation defined by $\kappa \approx 1$.) The solution with the '+' sign in the exponent corresponds to the solitonic solution, while the second one, with the '−' sign, can be treated as an antisolitonic solution. In order to demonstrate the relevance of the line-group method to the static solitons we rewrite the solution (3.2) in the form

$$\mathrm{e}^{(n/2)\vec{q}\cdot(\vec{z}-\vec{z}_0)} = \tan\left(\frac{n\varphi(\vec{z})}{4} - \frac{\pi}{4}\right). \tag{3.3}$$

When choosing the solitonic solution for the symmetry analysis, the application of the antisolitonic one leads to a mirror image of the solitonic solution in the ϕ–z plane, reflecting the mirror symmetry of soliton–antisoliton solutions. Let us apply a Seitz operator to both sides of equation (3.3); then we have the relation

$$\begin{aligned}\hat{D}(R|\vec{v}_R)\tan\left(\frac{n\varphi(\vec{z})}{4} - \frac{\pi}{4}\right) &= \tan\left(\frac{n\varphi((R|\vec{v}_R)^{-1}\vec{z})}{4} - \frac{\pi}{4}\right)\\ &= \tan\left(\frac{n\varphi((R^{-1}|-R^{-1}\vec{v}_R)\vec{z})}{4} - \frac{\pi}{4}\right)\end{aligned} \tag{3.4}$$

for the right-hand side, where R denotes the orthogonal transformation coupled to the elementary fractional translation of the given line group. According to the general rules concerning application of the symmetry operators, the transformed function on the left side of (3.3) can be written as

$$\begin{aligned}\hat{D}(R|\vec{v}_R)\mathrm{e}^{(n/2)\vec{q}\cdot(\vec{z}-\vec{z}_0)} \equiv \hat{D}(R|\vec{v}_R)\mathrm{e}^{(n/2)(\vec{q},\vec{z}-\vec{z}_0)} &= \mathrm{e}^{(n/2)(\vec{q},(R|\vec{v}_R)^{-1}(\vec{z}-\vec{z}_0))}\\ &= \mathrm{e}^{(n/2)(\vec{q},R^{-1}(\vec{z}-\vec{z}_0)-R^{-1}\vec{v}_R)} = \mathrm{e}^{(n/2)(R\vec{q},\vec{z}-\vec{z}_0-\vec{v}_R)}\end{aligned} \tag{3.5}$$

which becomes

$$\hat{D}(R|\vec{v}_R)\mathrm{e}^{(n/2)(\vec{q},\vec{z}-\vec{z}_0)} \equiv \mathrm{e}^{(n/2)(\vec{q},\vec{z}-\vec{z}_0)}\mathrm{e}^{(n/2)(\delta\vec{q},\vec{z}-\vec{z}_0)}\mathrm{e}^{-(n/2)(\vec{q}+\delta\vec{q},\vec{v}_R)} \tag{3.6}$$

using the expression $R\vec{q} = \vec{q} + \delta\vec{q}$, where $\delta\vec{q}$ describes the lattice modulation.

Assuming line-group invariance of the soliton lattice, we have that

$$\mathrm{e}^{(n/2)(\delta\vec{q},\vec{z}-\vec{z}_0)} = \mathrm{e}^{(n/2)(\vec{q}+\delta\vec{q},\vec{v}_R)}. \tag{3.7}$$

If we wish to explain the modulation of the ground-state lattice by elementary line-group symmetry, by suitable choice of the integration constant $\vec{z}_0 = -\vec{v}_R$ it is always possible to express the modulation of the 'primary' unperturbed crystal structure as

$$(\delta\vec{q}, \vec{z}) = (\vec{q}, \vec{v}_R) \tag{3.8}$$

i.e. by use of the elementary fractional translation $\vec{v}_R$ of a Seitz operator. The order of the screw axis, corresponding to the subgroup of generalized translations describing the static soliton lattice, can be determined very simply. With this aim, the periodicity of the tan function has been obtained from the transformation realized by the symmetry operator corresponding to an element $(P|\vec{v}_P)$ of the subgroup of generalized translations:

$$\hat{D}(P|\vec{v}_P)\tan\left[\frac{n}{4}\varphi(\vec{z}) - \frac{\pi}{4}\right] = \tan\left[\frac{n}{4}\varphi(P^{-1}(\vec{z}-\vec{v}_P)) - \frac{\pi}{4}\right] = \tan\left[\frac{n}{4}\varphi(\vec{z}) - \frac{\pi}{4} + k_P\pi\right] \tag{3.9}$$

where k_P denotes an integer. Performing a similar symmetry transformation by means of the element $(S|\vec{v}_S)$ of the same subgroup of generalized translations, and taking into account the assumed line-group invariance of the soliton structure, the difference of the arguments of the transformed functions can be calculated as

$$\varphi(S^{-1}(\vec{z}-\vec{v}_S)) - \varphi(R^{-1}(\vec{z}-\vec{v}_R)) = (k_S - k_R)\frac{4\pi}{n}. \tag{3.10}$$

This equality can be related to the phase-angle difference, which is obtained directly from the expression for the phase function (1.1) for two arbitrary layers (perpendicular to the direction of modulation) of the modulated structure:

$$\varphi_j - \varphi_i = \frac{\pi}{2}\bar{q}(j-i). \tag{3.11}$$

As is seen, formula (3.11) which originates from the line-group theory is in accordance with Bak's theory, having an arbitrary accuracy. The relevance of the line-group technique can be recognized in the case of non-linear excitations in magnetic chains, too. In the continuum approximation, the equation of motion corresponds to systems which can be described (Delhaes and Drillon 1987) in form of the sine–Gordon equation

$$\frac{\partial^2\vartheta}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2\vartheta}{\partial t^2} = m^2\sin\vartheta \tag{3.12}$$

where $\vartheta = \vartheta(z)$ denotes the angle between the magnetic field and the spin on the site with coordinate z and m is the characteristic length with

$$m^{-1} = a\sqrt{\frac{2JS}{g\mu_B H}}.$$

Also, S is the maximal projection of a spin on the direction of the external magnetic field, a is a constant and the characteristic velocity is

$$c = \frac{2aS\sqrt{AJ}}{\hbar}.$$

Similarly to the static solution (3.2) characteristic for the *static solitons*, the differential equation (3.12) has a non-linear character and a solution describing collective excitations, reflecting therefore *dynamic solitons* and meaning *de facto* moving domain walls in the same kink form as in the static case:

$$\vartheta(z,t) = 4\arctan\exp[\pm\gamma m(z - vt - z_0)] \tag{3.13}$$

where

$$\gamma = \sqrt{1 - \frac{v^2}{c^2}}.$$

In this way, the solution functions (3.2) and (3.13) describing static (topological) and dynamic solitons correspond to the simplest line-group symmetry transformations. It must nevertheless be emphasized that, although the mathematical formulae for these two types of soliton are formally analogous, their physical meanings are different. This difference and connection between them can be revealed by the application of the Goldstone's theorem (Goldstone 1961, Goldstone *et al* 1962). The solution (3.13) represents a more general character for investigation, as compared to the earlier procedures (Villain 1975, Davydov 1986), and so it has a wide scope for describing dynamic properties of solitons. In order to perform this analysis in the next section, we mention in advance here the important property of incommensurate structures that they are *continuously degenerate*, i.e. the thermodynamic potential is the same for each value of the phase angle.

4. Symmetry breaking and the cause of elementary excitations in incommensurate systems

For the symmetry analysis of the possible types of elementary excitation in the incommensurate crystals, first we point out the experimentally detectable continuous degeneracy of the ground state of incommensurate systems, ensuring the applicability of the Goldstone theorem. Having discussed the case of spontaneous symmetry breaking for continuously degenerate states and using local conservation laws (Wagner 1966), we analyse the basic symmetry properties of excitations existing in incommensurately modulated systems on the basis of the application of line-group technique. It results in gapless excitations and a generalization of the theory of symmetry breaking in discrete systems. According to this, the expression for the cross section for x-ray and neutron scattering relevant for Umklapp processes can be given as

$$\lim_{\vec{k}\to 0} F(\vec{k} + \vec{K}) \geqslant \frac{1}{k^2}\frac{(\vec{k}\cdot\vec{K}/k)^2 T\bar{\rho}^2_{\vec{K}}}{\gamma'} \tag{4.1}$$

where the quantity appearing in the argument of the function $F(\vec{k} + \vec{K})$ denotes the value of the impulse transfer in the actual inelastic scattering process and the overline on the Fourier coefficient $\rho_{\vec{K}}$ means the average value in quantum statistics. The fundamental expression (4.1) is based on Bogoliubov's quasi-average $1/k^2$ theorem, which describes the long-wavelength phonons and the gapless density excitations too, obeying the dispersion relation

$$\lim_{k\to 0} \omega(\vec{k} + \vec{K}) \propto k.$$

In the formula (4.1), $\gamma' = \gamma/(2nm)$ where n denotes the particle-number density, m is the mass of identical particles building up the whole system being examined, γ is a positive constant quantity, T represents the values of the absolute temperature and $\vec{K}$ is a vector from the reciprocal lattice, which brings the vector $\vec{k}$, corresponding to the final state of the scattering process, back to the first Brillouin zone. Because of this meaning, $\vec{K}$ consists of an integer

linear combination of generating elements (elementary vectors) of the reciprocal Bravais lattice. Therefore, symmetry analysis of the elementary excitations in incommensurate systems can be reduced to the calculation of the Fourier coefficients $\rho_{\vec{K}}$. That is, it is obvious from the Fourier expansion of the electron-density function

$$\rho(\vec{r}) = \sum_{\vec{k}} \rho_{\vec{k}} \mathrm{e}^{\mathrm{i}\vec{k}\cdot\vec{r}} \tag{4.2}$$

that its fundamental symmetry properties are also reflected in the Fourier coefficients in (4.2), as was also shown for discrete Q1D systems (Mészáros *et al* 1997, Kirschner *et al* 1998). On this supposition, it becomes possible to use equation (4.2) to describe elementary excitations of incommensurate systems easily, and classification of the selection rules can be explicitly incorporated into the expressions for the inelastic scattering cross section. According to the basic principles of quantum mechanics, the possible transition from a generally degenerate initial state $\{|i\rangle\}$ of a system to the final degenerate one $\{|f\rangle\}$ is characterized by the symmetry group G, its irrep $D^{(i)}(G)$ and the irrep $D^{(f)}(G)$. The selection rules can be determined by use of the direct product of representations, if we do not take into account the eventual accidental degeneracy and suppose that the irrep belonging to the perturbation-inducing transition is $D^{(\nu)}$. The transition becomes possible if the irrep corresponding to the final state is contained in the product $D^{(i)} \otimes D^{(\nu)}$. By use of the Clebsch–Gordan coefficients $C(i\nu|\mu)$, this product can be decomposed as follows:

$$D^{(i)} \otimes D^{(\nu)} = \sum_{i} \sum_{\nu} \sum_{\mu} C(i\nu|\mu) D^{(\mu)}. \tag{4.3}$$

In order to study the link between the Goldstone modes appearing in incommensurate systems (i.e. *phasons*) and special elementary excitations relevant for symmetries of Q1D systems, we here examine in detail selection rules which can be obtained using direct products of the irreps of line groups. As far as we are aware, the first such calculations were performed in the work of Damnjanović and Vujičić (1982), where the propagation of excitations along the main axis of helical systems has been demonstrated, obeying the conservation laws of the quasi-impulse and the quasi-angular momentum. For the sake of simplicity, in the present section we will examine possible elementary excitations of non-magnetic incommensurate systems, also having helical symmetry. Since the symmetry group of helical systems can be identified as the simplest line group, $Ln_p = (n/r) \otimes \mathrm{C}_q^a$, the selection rules that are suitable for such systems can be derived by use of irreps of the group n/r of generalized translations and irreps of the point group C_q^a. If the distance between nearest-neighbour scattering centres in a helical system is ζ, then the irreps of the subgroup of generalized translations n/r are of the form

$$D^{(\mu)}(R|\nu_R) \equiv {}_kA((\mathrm{C}_n^r|\nu)^t) = \mathrm{e}^{\mathrm{i}k\zeta} \qquad k \in \left(-\frac{\pi}{\zeta}, \frac{\pi}{\zeta}\right] \tag{4.4}$$

where t is an integer and the index k denotes a *de facto* wave vector, whose possible values (in accordance with the Born–Kármán cyclic boundary conditions for Q1D systems (Božović *et al* 1978, Božović and Vujičić 1981)) also denote the non-equivalent irreps of n/r. Since irreps of the cyclic point groups are known, and irreps of the second constitutive symmetry group of the helical system are one dimensional, the general form of the matrices, which represent general elements of the full symmetry group Ln_p, is

$${}_kA_m((\mathrm{C}_n^r|n)^t\mathrm{C}_q^s) = \mathrm{e}^{\mathrm{i}kt\zeta}\mathrm{e}^{\mathrm{i}msa} \tag{4.5}$$

where $\mathrm{e}^{\mathrm{i}ms\alpha} = A_m(\mathrm{C}_q^s)$ denotes the one-dimensional matrix corresponding to the irrep of the cyclic discrete rotational group and the selection rules can be presented as

$$\Delta k \equiv k_f - k_i = k + \frac{2j\pi}{\zeta} \qquad \Delta m \equiv m_f - m_i = m + zq \tag{4.6}$$

where s, j and z are integers. This result originated from purely mathematical calculations by Damnjanović and co-workers (Damnjanović *et al* 1983, Damnjanović and Božović 1984). The first relation of (4.6) expresses the conservation of the quasi-impulse, while the second one corresponds to the conservation of the quasi-angular momentum. It can be immediately seen that these selection rules describe exactly the earlier-mentioned elementary excitations called phasons. Therefore, these selection rules can be obtained on the basis of (4.3), as

$$\vec{k}_f = \vec{k}_i + \vec{k}_v \pm \vec{K} \tag{4.7}$$

for Umklapp processes and the restrictions being satisfied simultaneously are

$$m_f = m_i + m_v \pm p \tag{4.8}$$

following from the selection rules concerning quasi-angular momenta, where the values of p must be taken from the second relation in (4.6). Relation (4.8) extends the mathematical formalism of the x-ray and neutron scattering from modulated crystals in a remarkable manner, since it represents selection rules which, as far as we are aware, have not been applied to evaluate experimental data in such scattering experiments. By use of this last restriction, we give completely new expressions for the relevant inelastic scattering cross sections and energies of the elementary excitations in incommensurate systems. That is, the description of the spontaneous symmetry breaking of continuous symmetries proposed by Wagner can be directly connected to the selection rules (4.6). For this, it is only necessary to take $\vec{K}$ appearing in (4.1) from (4.7) and to require at the same time the validity of equation (4.8). In order to illustrate the completeness of the line-group-technique-based theory of symmetry-breaking phenomena in modulated systems, we recall the result published in the paper by Milošević and Damnjanović (1993). According to this result, the Jahn–Teller theorem (Jahn and Teller 1937) remains valid for infinite Q1D systems too, as was demonstrated originally for finite 3D molecules.

Finally, all partial results discussed in the present paper can be generalized by means of the Goldstone theorem applied to the incommensurately modulated crystals as follows. Let the high- and low-temperature symmetry groups (usually corresponding in turn to the higher- and lower-symmetry states of the system) be denoted by G_H and G_L, respectively. According to the general concept (Mermin 1979) for states with broken symmetry, G_L is a subgroup of G_H (i.e. $G_L < G_H$) and we have the following coset decomposition:

$$G_H = G_L + t_2 G_L + \cdots + t_s G_L \qquad s = \frac{|G_H|}{|G_L|} \tag{4.9}$$

where the coset representatives t_s are the symmetry elements lost during the phase transition and the symmetries of propagating elementary excitations appearing after symmetry breaking, whereas the symbol $|\cdots|$ denotes the order of the group. The general formula (4.9) can be refined as follows. Following the method of Izyumov and Syromyatnikov (1990), in the case of ordinary 3D crystals studied within SFS theory, the lost symmetry elements could be point-group elements (giving *orientational domains* in the low-temperature phase), translations (resulting similarly in *antiphase domains*), or products of such lost orthogonal and translational symmetry elements. All of these cases can be directly described by use of (4.9). Also, the relationship (4.9) is valid in the case of continuous groups and the coset representatives may even form a convergent sequence of group elements (i.e. we can speak about a convergent sequence of cosets, too). The existence of chirality domains in 'helimagnetic' crystal samples was experimentally confirmed by the neutron topography technique (Baruchel 1994). Such domains cannot be simply classified within framework of the SFS theory. Due to their above-mentioned richer algebraic structure and the natural appearance of symmetry elements with

non-crystallographic character, line groups are more suited for describing of the symmetry-breaking phenomena in modulated crystals. Since relations (3.6)–(3.10) describe the simplest possible soliton lattices only, whose domain structures are described using Seitz operators of subgroups of generalized translations of relevant line groups, the generalization explained by the following postulate is straightforward. In the case of breaking of continuous symmetry, taking place at the appearance of phasons, the coset representatives t_s are composed from symmetry elements g_c belonging to a continuous group and line-group elements corresponding to symmetry operations describing mutual space and time relations of domains, i.e. we have $t_s = g_c(P|\vec{t})$ with a symmetry transformation from a line group and denoted by the Coster symbol $(P|\vec{t})$.

5. Formulae for the Debye–Waller factor in the case of incommensurate structures

In the following we apply the earlier-discussed mathematical formalism in order to calculate the actual relations of Debye–Waller factors for the case of crystals modulated incommensurately. According to the kinematic approximation of the theory of scattering of x-rays or neutrons by crystals, the Debye–Waller factors describing the influence of lattice vibrations on the diffracted intensities must be calculated according to the formula $\langle e^{i\vec{\kappa}\cdot\vec{u}_{nj}}\rangle$, where $\vec{u}_{nj}$ is the displacement of the jth scattering centre in the nth unit cell and the symbol $\langle\cdots\rangle$ denotes the quantum-statistical average. Then, the Debye–Waller factors enter generally the expressions for the scattering cross section as $e^{-\langle(\vec{\kappa}\cdot\vec{u}_{nj})^2\rangle}$ (Cowley 1975). The displacements of the atomic scattering centres with respect to their positions in the absence of modulation are given by the functions

$$\vec{u}_{nj} = \vec{u}_j \sin(\vec{k}_{mod}\cdot\vec{R}_n + \psi_j) \tag{5.1}$$

where the wave vector $\vec{k}_{mod}$ describes the modulation and corresponds to the reciprocal-space vector $\delta\vec{q}$ applied in section 3. Using the expansion

$$e^{iz\sin\alpha} = \sum_{m=-\infty}^{+\infty} J_m(z)e^{im\alpha}$$

the exponential factor necessary for calculation of the Debye–Waller factor can be written as

$$e^{i\vec{\kappa}\cdot\vec{u}_{nj}} = \sum_{m=-\infty}^{m} J_m(\vec{\kappa}\cdot\vec{u}_j)e^{im(\vec{k}_{mod}\cdot\vec{R}_n+\psi_j)}. \tag{5.2}$$

Employing conventional formulae applied in diffraction physics, the intensities of the Bragg reflections as well as their positions in the reciprocal space can be determined by the usual Fourier-transformation procedure, which yields the result

$$I_r(\vec{\kappa}) = 8\pi^3\frac{N}{V_c}|f_m|^2\delta(\vec{\kappa}-\vec{G}+m\vec{k}_{mod}) \qquad \left(f_m = \sum_{j=1}^{\nu} f_j e^{i\vec{\kappa}\cdot\vec{R}_j} J_m(\vec{\kappa}\cdot\vec{u}_j)e^{im\psi_j}\right) \tag{5.3}$$

(Overhauser 1971, Giuliani and Overhauser 1981). Although structures with multiple modulation also exist (Bruce and Cowley 1981, Izyumov and Syromyatnikov 1990), we limit our investigation at present to the case of simple modulation. (It should be noted that multiply modulated structures can also be treated by the line-group technique by using the concept of the *generalized semi-direct product* of line groups (Kirschner *et al* 1997), and generalization of the following calculations to the more complicated structures is straightforward.) Then, in the approximation proposed by Overhauser, the Debye–Waller factor can be written as

$$e^{-W_j(m)} = \left\langle e^{im\,\delta\psi_j}\frac{J_m[\vec{\kappa}\cdot(\vec{u}_j+\delta\vec{u}_j)]}{J_m(\vec{\kappa}\cdot\vec{u}_j)}\right\rangle. \tag{5.4}$$

If the fluctuations of the amplitude and phase satisfy

$$m^2\langle(\delta\psi_j)^2\rangle \ll 1 \qquad m^2\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle \ll 1$$

the following approximate expression can be obtained for the Debye–Waller factors:

$$W_j(m) \approx \frac{m^2}{2}\langle(\delta\psi_j)^2\rangle - \frac{|m|}{2}(|m|-1)\frac{\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle}{(\vec{\kappa}\cdot\vec{u}_j)^2}. \tag{5.5}$$

Thus, the refinement of the scattering formalism concerning incommensurate structures can be performed on the basis of line-group theory in order to simplify the formulae (5.4) and (5.5) using the actual generating elements.

Table 1 contains the formulae describing actual Debye–Waller factors allowed by line-group symmetries and derived according to the method just explained. In order to give information concisely, we do not list all of the generating elements of the line groups—only their isogonal point groups as well as the generating elements of the subgroups of generalized translations, appearing in the richest algebraic factorizations. When subgroups of generalized translations are generated by purely integer translations, the generating elements are also not indicated.

According to these results, it is obvious that expression (5.5) obtained for Debye–Waller factors for incommensurate structures may have at least three different forms for symmetry reasons. They can be used directly in the algorithms necessary for calculations in the structure determination of crystals required by diffraction experiments.

6. Conclusions

(1) The line-group method is able to make a clear distinction between Q1D and strictly 1D systems and therefore can be used to perform a further refinement of theoretical descriptions of some basic phenomena in the physics of condensed matter related to the properties of low-dimensional subsystems (such as the theory of Peierls transitions) via symmetry analysis based on line-group methods. Comparison of the Fourier formalism developed to meet the needs of the structural investigation of stereoregular polymers by means of x-rays and the relevant formalism elaborated within the framework of N-dimensional crystallography indicates that structure analysis of incommensurately modulated crystals can be based on the line-group technique as well. In particular, Debye–Waller factors can be simplified directly by using this method, which provides their actual values. Also, the line-group technique gives a very simple theory for general treatment of the symmetry-breaking phenomena in the incommensurately modulated crystals. Application of the same technique may lead to a refined description and deeper understanding of the already-described soliton-type elementary excitations in HTSs, present according to the latest experiments. It has been shown that Q1D subsystems in HTS materials can play an important role in the HTS mechanism. This new result demands the re-examination of the concept of dynamic solitons in non-magnetic and/or magnetic systems, and further investigation of striped phases may represent a useful research topic in the future.

(2) As a continuation of this work, performing a generalization of the description of vibronic interactions in condensed matter systems of different types is immediately possible. The investigation of the similar effect of the Jahn–Teller phenomenon and Peierls instability in the creation of superconductivity by the line-group method seems also to be reasonable.

Table 1. Line groups, their generating elements and the relevant Debye–Waller factors.

	Symbol	Generating element	Debye–Waller factor
1	Lq_p	C_q, $(C_q^r\|1/q)$	$\frac{(mq)^2}{2}\langle(\delta\psi_j)^2\rangle - \frac{\|mq\|}{2}(\|mq\|-1)\frac{\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle}{(\vec{\kappa}\cdot\vec{u}_j)^2}$
2	$L(\overline{2n})$, $L\tilde{n}$	S_{2n}	$2m^2n^2\langle(\delta\psi_j)^2\rangle - \|mn\|(\|2mn\|-1)\frac{\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle}{(\vec{\kappa}\cdot\vec{u}_j)^2}$
3	$L(\overline{2n})$, Ln/m	$C_{n\mathrm{h}}$	$\frac{(mn)^2}{2}\langle(\delta\psi_j)^2\rangle - \frac{\|mn\|}{2}(\|mn\|-1)\frac{\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle}{(\vec{\kappa}\cdot\vec{u}_j)^2}$
4	$L(2n)_n/m$	$C_{2n\mathrm{h}}$, $(\sigma_\mathrm{v}\|1/2)$	$2m^2n^2\langle(\delta\psi_j)^2\rangle - \|mn\|(\|2mn\|-1)\frac{\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle}{(\vec{\kappa}\cdot\vec{u}_j)^2}$
5	Lq_p22, Lq_p2	D_q, $(C_q^r\|1/q)$	$\frac{(mq)^2}{2}\langle(\delta\psi_j)^2\rangle - \frac{\|mq\|}{2}(\|mq\|-1)\frac{\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle}{(\vec{\kappa}\cdot\vec{u}_j)^2}$
6	$Lnmm$, Lnm	$C_{n\mathrm{v}}$	$\frac{(mn)^2}{2}\langle(\delta\psi_j)^2\rangle - \frac{\|mn\|}{2}(\|mn\|-1)\frac{\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle}{(\vec{\kappa}\cdot\vec{u}_j)^2}$
7	$Lncc$, Lnc	$C_{n\mathrm{v}}$, $(\sigma_\mathrm{v}\|1/2)$	$\frac{(mn)^2}{2}\langle(\delta\psi_j)^2\rangle - \frac{\|mn\|}{2}(\|mn\|-1)\frac{\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle}{(\vec{\kappa}\cdot\vec{u}_j)^2}$
8	$L(2n)_nmc$	$C_{2n\mathrm{v}}$, $(C_q^r\|1/q)$	$2m^2n^2\langle(\delta\psi_j)^2\rangle - \|mn\|(\|2mn\|-1)\frac{\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle}{(\vec{\kappa}\cdot\vec{u}_j)^2}$
9	$L(\overline{2n})2m$, $L\tilde{n}m$	$D_{n\mathrm{d}}$, $(\sigma_\mathrm{v}\|1/2)$	$\frac{(mn)^2}{2}\langle(\delta\psi_j)^2\rangle - \frac{\|mn\|}{2}(\|mn\|-1)\frac{\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle}{(\vec{\kappa}\cdot\vec{u}_j)^2}$
10	$L(\overline{2n})2c$, $L\tilde{n}c$	$D_{n\mathrm{d}}$, $(\sigma_\mathrm{v}\|1/2)$	$\frac{(mn)^2}{2}\langle(\delta\psi_j)^2\rangle - \frac{\|mn\|}{2}(\|mn\|-1)\frac{\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle}{(\vec{\kappa}\cdot\vec{u}_j)^2}$
11	Ln/mmm, $L(\overline{2n})2m$	$D_{n\mathrm{h}}$, $(\sigma_\mathrm{v}\|1/2)$	$\frac{(mn)^2}{2}\langle(\delta\psi_j)^2\rangle - \frac{\|mn\|}{2}(\|mn\|-1)\frac{\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle}{(\vec{\kappa}\cdot\vec{u}_j)^2}$
12	Ln/mcc, $L(\overline{2n})2c$	$D_{n\mathrm{h}}$, $(\sigma_\mathrm{v}\|1/2)$	$\frac{(mn)^2}{2}\langle(\delta\psi_j)^2\rangle - \frac{\|mn\|}{2}(\|mn\|-1)\frac{\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle}{(\vec{\kappa}\cdot\vec{u}_j)^2}$
13	$L(2n)_n/mcm$	$D_{2n\mathrm{h}}$, $(\sigma_\mathrm{v}\|1/2)$	$2m^2n^2\langle(\delta\psi_j)^2\rangle - \|mn\|(\|2mn\|-1)\frac{\langle(\vec{\kappa}\cdot\delta\vec{u}_j)^2\rangle}{(\vec{\kappa}\cdot\vec{u}_j)^2}$

References

Bak P 1981 *Phys. Rev. Lett.* **46** 791

Baruchel J 1994 *Neutron and Synchrotron Radiation for Condensed Matter Studies* vol 1, ed J Baruchel, J L Hodeau, M S Lehmann, J R Regnard and C Schlenker (Berlin: Springer) pp 399–409

Baudour J L and Sanquer M 1983 *Acta Crystallogr.* B **39** 75

Božović I B and Vujičić M 1981 *J. Phys. A: Math. Gen.* **14** 777

Božović I B, Vujičić M and Herbut F 1978 *J. Phys. A: Math. Gen.* **11** 2133

Bruce A D and Cowley R A 1978 *J. Phys. C: Solid State Phys.* **11** 3609

Bruce A D and Cowley R A 1981 *Structural Phase Transitions* (London: Taylor and Francis)

Cowley J M 1975 *Diffraction Physics* (Amsterdam: North-Holland)

Cowley R A and Bruce A D 1978 *J. Phys. C: Solid State Phys.* **11** 3577

Cummins H Z 1990 *Phys. Rep.* **185** 211
Dai P, Mook H A, Aeppli G, Hayden S M and Doğan F 2000 *Nature* **406** 965
Damnjanović M and Božović I B 1984 *J. Phys. A: Math. Gen.* **17** 747
Damnjanović M, Božović I B and Božović N 1983 *J. Phys. A: Math. Gen.* **16** 3937
Damnjanović M, Milošević I, Vuković T and Sredanović R 1999a *J. Phys. A: Math. Gen.* **32** 4097
Damnjanović M, Milošević I, Vuković T and Sredanović R 1999b *Phys. Rev.* B **60** 2728
Damnjanović M and Vujičić M 1982 *Phys. Rev.* B **25** 6987
Davydov A S 1986 *Ann. Phys., Lpz.* **43** 93
Delhaes P and Drillon M (ed) 1987 *Organic and Inorganic Low Dimensional Crystalline Materials (NATO ASI Series B)* (New York: Plenum) p 168
de Wolff P M 1974 *Acta Crystallogr.* A **30** 777
de Wolff P M 1977 *Acta Crystallogr.* A **33** 493
de Wolff P M, Janssen T and Janner A 1981 *Acta Crystallogr.* A **37** 625
Dzyaloshinskii I E 1964 *Sov. Phys.–JETP* **19** 960
Dzyaloshinskii I E 1965a *Sov. Phys.–JETP* **20** 223
Dzyaloshinskii I E 1965b *Sov. Phys.–JETP* **20** 665
Giuliani G F and Overhauser A W 1981 *Phys. Rev.* B **23** 3737
Goldstone J 1961 *Nuovo Cimento* **19** 154
Goldstone J, Salam A and Weinberg S 1962 *Phys. Rev.* **127** 965
Hermann C 1929 *Z. Kristallogr.* **69** 250
Hornreich R M, Luban M and Shtrikman S 1975 *Phys. Rev. Lett.* **35** 1678
Izyumov Yu A and Syromyatnikov V N 1990 *Phase Transitions and Crystal Symmetry* (Dordrecht: Kluwer)
Jahn H A and Teller E 1937 *Proc. R. Soc.* A **161** 220
Kirschner I, Mészáros Cs and Laiho R 1997 *Z. Phys.* B **104** 289
Kirschner I, Mészáros Cs and Laiho R 1998 *Eur. Phys. J.* B **2** 191
Mermin N D 1979 *Rev. Mod. Phys.* **51** 591
Mermin N D 1992 *Rev. Mod. Phys.* **64** 3
Mészáros Cs, Bálint Á, Kirschner I, Laiho R, Lähderanta R and Bánkuti J 1997 *Superlatt. Microstruct.* **21** 381
Mészáros Cs and Bánkuti J 1994 *Phys. Status Solidi* b **183** 73
Milošević I and Damnjanović M 1993 *Phys. Rev.* B **47** 7805
Mook H A, Dai P, Doğan F and Hunt R D 2000 *Nature* **404** 729
Mook H A and Doğan F 1999 *Nature* **401** 145
Overhauser A W 1971 *Phys. Rev.* B **3** 3173
Peierls R 1955 *Quantum Theory of Solids* (Oxford: Oxford University Press)
Pouget J P 1994 *Neutron and Synchrotron Radiation for Condensed Matter Studies* vol 2, ed J Baruchel, J L Hodeau, M S Lehmann, J R Regnard and C Schlenker (Berlin: Springer) pp 245–60
Vainshtein B K 1966 *Diffraction of X-rays by Chain Molecules* (Amsterdam: Elsevier)
Villain J 1975 *Physica* B + C **79** 1
Vujičić M, Božović I B and Herbut F 1977 *J. Phys. A: Math. Gen.* **10** 1271
Wagner H 1966 *Z. Phys.* **195** 273
Zaanen J 2000 *Nature* **404** 714