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Theory of Phasons in Aperiodic Crystals

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The possibility of embedding a quasi-periodic structure into a higher-dimensional superspace gives rise to an interpretation of certain dynamical excitations as motions in the additional space. These excitations then are called phason excitations. There is always infinite degeneracy of the ground state of an aperiodic quasi-periodic system, but this degeneracy leads only to zero frequency modes if the atomic surfaces are smooth. If this is not the case, there is a phason gap.

When the atomic surfaces are continuous, the linear phason excitations with low frequency may get an arbitrary amplitude. The excitations are almost frictionless when the velocity is below a threshold value. Above that value there is dissipation (i.e. energy transfer to phonons) even for smooth atomic surfaces.

Keywords Aperiodic crystals; phasons; sliding modes

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1. Introduction

Quasiperiodic crystals may be embedded into a higher-dimensional space. So it seems that in principle the number of degrees of freedom is larger than for physical space, periodic crystals. This is not true because the total number of degrees of freedom is always the number of particles times the dimension of physical space. Nevertheless some of the low-energy modes may have a simpler interpretation as motions in the higher-dimensional space. These phason modes are not additional but are related to a degeneracy of the ground state.

According to the definition, quasi-periodic crystals have a diffraction pattern with sharp spots on positions belonging to a Fourier module:

$$\vec{k} = \sum_{i=1}^{n} h_i \vec{a}_i^*.$$

The number of rationally independent basis vectors is the rank of the module. Modulated phases, composites and quasi-crystals are examples of quasi-periodic crystals. Modulated structures have a lattice periodic basic structure and a periodic displacement from these basic structure positions. A simple case of rank four has positions $\vec{n} + \vec{f}(\vec{q} \cdot \vec{n})$, with a periodic function $\vec{f}$ with period 1 and $\vec{q} \cdot \vec{n} \neq 0(mod 1)$, $\forall \vec{n} \in \mathcal{L}$, $\mathcal{L}$ being the 3D Bravais lattice. A composite has two or more subsystems which are modulated structures with mutually incommensurate basic structures. A simple model has two subsystems. The atoms of the first subsystem are at $\vec{n} + \vec{f}(\vec{e} \cdot \vec{n})$, $\vec{n} \in \mathcal{L}$, those of the second subsystem ar $\vec{m} + \vec{g}(\vec{e} \cdot \vec{m})$, $\vec{m} \in \mathcal{L}'$, the two Bravais structures $\mathcal{L}$, $\mathcal{L}'$ have the same basis vectors in the x and y directions, and

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incommensurate lattice constants a and b in the z direction ($\vec{e}$ is a unit vector in the z direction). Finally the functions $\vec{f}$ and $\vec{g}$ are periodic with periods b and a, respectively.

2. Embedding

Quasiperiodic structures are the intersection of a lattice periodic structure in a space of dimension, equal to the rank of the Fourier module, and the physical space. The Fourier module is then the projection of the reciprocal lattice of the lattice periodic structure. Another important fact is that for incommensurate structures the projection of the direct lattice on the internal space is dense.

For modulated phases and composites the result of the embedding is as follows. For the modulated phase, as given above, the embedding is

$$(\vec{n} + \vec{f}(\vec{q} \cdot \vec{n} + t),\ t), \tag{2.1}$$

The embedding is invariant under the lattice vectors in x and y direction and under the translations $(\vec{c}, -\vec{q} \cdot \vec{c})$ and $(0, 1)$.

For the composites the embedding is (see Fig. 1)

$$\begin{gathered}(\vec{n} + \vec{f}(\vec{e} \cdot \vec{n} + t) + Z_1 t\vec{e}, t) \\ (\vec{m} + \vec{g}(\vec{e} \cdot \vec{m} - t) - Z_2 t\vec{e}, t)\end{gathered} \tag{2.2}$$

where $Z_1 + Z_2 = 1$. (The periods of $\vec{f}$ and $\vec{g}$ are b and a, resp.). This embedded structure is invariant under the translations in x and y direction, and under translations $(Z_2 a\vec{e}, -a)$ and $(Z_1 b\vec{e}, b)$.

Because $\vec{q} \cdot \vec{c}$ and a/b are irrational, the projection of the lattice on the internal space is dense in the zt-plane.

The embedded structures consist of periodic arrays of wavy lines, the atomic surfaces. If the modulation functions are continuous the atomic surfaces are without boundaries. However, when the modulation functions are discontinuous, the atomic surfaces consist of disjunct pieces. This is similar to the situation in quasi-crystals where the atomic surfaces are finite $(n - 3)$-dimensional objects in n dimensions.

Under an n-dimensional lattice translation the mutual distances of the particles are constant. Therefore, there is a dense set of values of t for which the potential energy of

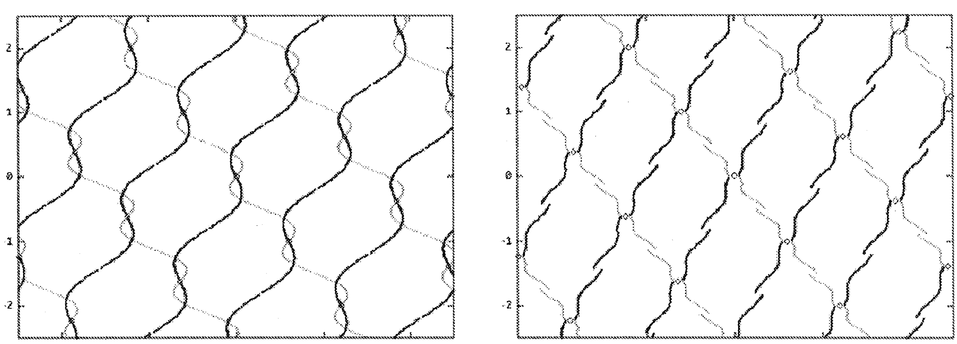

FIGURE 1 Embedding of an incommensurate composite into superspace for (a) continuous modulation, and (b) discontinuous modulation.

the three-dimensional structure for an intersection at this t is the same as for $t = 0$. This means that the ground state in infinitely degenerate in the variable t. Moreover, when the atomic surfaces are smooth the potential energy is invariant under arbitrary shifts of the coordinate t.

3. Dynamical Symmetry

The same dynamical symmetry may be seen as follows. Consider an incommensurate modulated phase with positions of the atoms at $x_n = na + f(na)$ with $f(x) = f(x+b)$. A shift $x_n \to x_n + \delta$ will keep all inter-atomic distances invariant. Therefore, it is a symmetry for the Hamiltonian and it leads to the existence of a zero frequency mode, the $k = 0$ acoustic mode. The transformation $x_n \to x_{n+p} - pa$ leads to positions $na + f(na + pa - qb) = x_n + f(na + \epsilon)$. For every value of ϵ there are integers p and q such that $pa - qb$ approximates ϵ arbitrarily well. This means that there is a dense set of shifts of the phase of the modulation function which leave the potential energy invariant. The implication is that there is for smooth modulation function a zero frequency mode which can be seen as a shift of the phase, a phason.

For incommensurate composites similar dynamic symmetries exist. If the atoms of two subsystems are located at $x_n = x_0 + na + f(na)\,(f(x) = f(x+b))$, and $y_m = y_0 + mb + g(mb)\,(g(y) = g(y+a)$ a first dynamical symmetry consists of the shifts $x_n \to x_n + \delta$ and $y_m \to y_m + \delta$. A second transformation is $x_n \to x_{n+p} - pa$ and $y_m \to y_{m+q} - pa$. Using the periodicity of the modulation functions this gives $x_n \to x_0 + na + f(na+\epsilon)$ and $y_m \to y_0 + mb - \epsilon + g(mb - \epsilon)$ with $\epsilon = pa - qb$. Therefore the Hamiltonian is invariant under a dense set of phase shifts of the modulation functions. For continuous modulation functions this will lead again to an additional zero frequency phason mode.

4. Phasons

Because for incommensurate modulated phases a shift in internal space is just a shift in the argument (the phase) of the modulation function, the zero frequency infinite wave-length and similar long wave-length excitations were called phase excitations or phasons. For incommensurate composites such an excitation not only shifts the phase but also the relative positions of the centres of mass of the subsystems. Therefore, these were usually called sliding modes. Finally for similar phenomena in quasi-crystals the same word 'phason' was used, but now with at least three different meanings. Analogous to the first two types a similar shift in internal space for a quasi-crystal leaves the Hamiltonian invariant for a dense set of shift values. However, now the displacements cannot be made arbitrarily small. There are local shifts leading to a finite jump of a particle, a phason jump. If the jumps are collective in the sense that they are induced by a long wave-length oscillation of the physical space, there are collective phasons. Finally the word phason in used for a strain of the higher-dimensional lattice which effects only the internal components. The three types of 'phason' have been seen experimentally. Phason jumps have been seen by time-of-flight neutron scattering [1] and time resolved electron microscopy [2]. Phason strain can often be seen in HREM pictures as discontinuities in rows of particles. Collective phason excitations have been reported by de Boissieu [3]. They are always diffusive, because of the jumps. Here we shall limit ourselves to incommensurate modulated phases and composites.

Dynamical excitations described by a (uniform or long wave-length) displacement δ are acoustic phonons. Excitations described as (uniform or long wave-length) shift of the internal coordinate are called phasons. In principle, they are oscillations around the equilibrium positions, and for that reason phonons, but they have a special character. A pure infinite wave-length phason is a phonon with an eigenvector proportional to the derivative of the modulation function.

5. Phason Content of Phonons

Traditionally, low-energy modes in quasi-periodic crystals are thought to be either acoustical phonons or phasons. In fact these correspond to special polarizations in the embedding space. Other polarizations correspond to phonons that have various phason contents.

For an embedded IC modulated structure $(na + f(na + t), t)$ a displacement with polarisation vector

$$P = \epsilon(\cos\theta, \sin\theta)$$

has the displacement field in physical space given by

$$x_n \to x_n + u_n, \quad u_n = \epsilon[\cos(\theta) + \sin(\theta) f'(na)].$$

Then

$$U_1 = \epsilon \cos(\theta) = \frac{1}{N} \sum_n u_n$$
$$U_2 = \frac{1}{N} \sum_n \frac{u_n - U_1}{f'(na)},$$

from which follow ϵ and θ by

$$\tan(\theta) = U_2/U_1$$
$$\epsilon = \sqrt{U_1^2 + U_2^2}.$$

For a pure acoustic phonon $u_n = \delta, U_1 = \delta, U_2 = 0, \theta = 0$. The polarisation vector lies in the physical space. For a pure phason $u_n = \epsilon f'(na)$, and $U_1 = 0, U_2 = \epsilon, \theta = \pi/2$. Then the polarisation lies in the internal space. We argued [7] that in modulated phases, under rather general conditions, phonons and phasons do not mix and therefore the possible polarisations are restricted to $\theta = 0, \pi/2$.

For an incommensurate composite shift parameters Δ_i are defined by

$$\triangle_1 = \frac{1}{N_1} \sum_n u_n, \quad \triangle_2 = \frac{1}{N_2} v_m,$$

where N_i is the number of particles in subsystem i, and u_n and v_m the eigenvector displacements of the phonon in the two chains. Then

$$\triangle_1 = \epsilon(\cos(\theta) + Z_1 \sin(\theta))$$
$$\triangle_2 = \epsilon(\cos(\theta) - Z_2 \sin(\theta)).$$

Consequently, the quantities ϵ and θ follow from

$$\tan(\theta) = \frac{a \sum_n u_n - b \sum_m u_m}{a Z_2 \sum_n u_n + b Z_1 \sum_m u_m}$$
$$\epsilon = \frac{1}{Z_1 + Z_2}((\triangle_1 - \triangle_2)\sin(\theta) + (Z_2 \triangle_1 + Z_1 \triangle_2)\cos(\theta)).$$

In this way the phason content, parametrised by θ, is calculated for a phonon with displacements (u_n, v_m). Contrary to the case of modulated phases, there is no a priori restriction on the value of θ. This can even change within a phonon branch [7]. Again a pure acoustic phonon has $\theta = 0$. A pure phason is such that it preserves the mass center ($\rho_1\triangle_1 + \rho_2\triangle_2 = 0$) therefore has $\tan\theta = \frac{\rho_1+\rho_2}{\rho_2 Z_2 - \rho_1 Z_1}$.

For a phonon with wave vector k the summations over the particles get a weight factor $\exp(ikna)$ for an IC phase and a similar adaptation for an incommensurate composite. In that way, even if all excitations are considered as phonons their phason content can be calculated using the phonon eigenvectors.

6. Phason Dynamics

The Hamiltonian of a crystal is invariant under a uniform shift. This implies that there is momentum conservation. A crystal set into motion will keep its initial velocity. A pure phason for a quasi-periodic structure with continuous modulation function is a phonon that can be seen as a uniform shift of the phase of the modulation. Although the ground state is infinitely degenerate, the equations of motion become non-linear if the displacements are no longer infinitesimal. Kinetic effects then may play a role, even if the modulation function is smooth. To check whether there is energy loss when the amplitudes are so big that non-linear terms become important, numerical calculations are needed. We shall study phasons in IC modulated phases and sliding modes in composites.

For an IC phase we take as model the discrete frustrated Φ^4 (DIFFOUR) model [4]. It is a tetragonal lattice with particles with one degree of freedom per vertex and a Hamiltonian

$$H = \sum_n \left(\frac{p_n^2}{2} - \frac{a}{2} y_n^2 + \frac{1}{4} y_n^4 + \sum_{n'} y_n y_{n'} + d \sum_{n''} y_n y_{n''} \right) \tag{6.3}$$

a characterises the site potential and therefore the displacive ($a \approx 0$) order-disorder ($a \to \infty$) character of the ground state. The parameter d characterises the frustration between the nearest neighbour and the next-nearest-neighbour interactions. For a fixed value of d, the ground state was calculated for a series of values of a. For $d = -0.35$ there is a critical value a_i below which the ground state is the para-phase. Above a_i the ground state is a modulated phase with an almost sinusoidal modulation function. For still higher values the modulation function squares up and between 0.5 and 0.6 there is a transition from a continuous modulation function to a discontinuous modulation function. The discontinuities increase in size, and above a value a_c the ground state is a superstructure.

In the region between a_i and the value a_d where the discontinuities set in, there is a zero frequency mode, with an eigenvector which is the derivative of the modulation function. Therefore, this mode can be interpreted as the phason mode. However, above a_d a phason gap opens up.

For incommensurate composites the situation is similar, as follows from numerical calculations in the double chain model [5–7]. When the modulation function is smooth, there is a zero frquency mode. From the integration of the non-linear equations of motion it follows that when the initial velocity is below a threshold value, then the subsystems slide over each other without friction. When the velocity is higher than the threshold value, there is energy transfer from the relative center of mass motion to phonons. If the modulation function is discontinuous, the sliding motion is always dissipative. The energy is distributed over all phonon modes.

7. Concluding Remarks

We have studied the phason dynamics of quasi-periodic crystals. The following conclusions can be drawn:

- Quasi-periodic crystals have an infinitely degenerate ground state.
- If the modulation functions are smooth, in other words if the atomic surfaces are simply connected, there is a zero frequency mode, called the phason mode for modulated phases and the sliding mode for composites.
- All phonons may be characterised by their phason content, an angle variable equal to 0 for the pure phonon. The angle gives the orientation of the polarisation vector in superspace.
- For large amplitude zero frequency phasons go over into solitary phason waves.
- The solitary phason waves have a very low damping if the initial velocity is below a critical value.
- When the initial velocity exceeds this critical value, the non-linear motion becomes dissipative, and the propagation stops.

The fact that for real quasi-periodic crystals zero frequency phasons have seldom been found, is possibly due to the presence of defects.

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