Ground State and Phonon Excitations in Aperiodic Composites

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Abstract

The phase diagram in parameter space of the Double Chain Model for the existence of the phason gap is calculated. Results for different interchain potentials are compared. The modulation function and the phason mode are studied.

1. Introduction

Incommensurate composites belong to the class of aperiodic crystals. They consist of at least two interpenetrating crystalline subsystems. In one or more crystallographic directions the periodicities of these subsystems are incommensurate, i.e. the ratio of the periodicities is not a rational number.

An example is the incommensurate inclusion compound C_nH_{2n+2}/urea. The urea molecules (the hosts) form a hexagonal lattice, with the alkane molecules (the guests) situated in the channels of this lattice. Theoretically, these incommensurate systems show the existence of a sliding or phason mode for not too strong interchain coupling. However, experiments[1-3] are still not conclusive on the existence of this mode in alkane/urea systems.

The recently introduced Double Chain Model[4] provides a suitable model to investigate inclusion composites. It takes into account the discreteness of the real crystal. It consists of two mutually incommensurate chains that interact with each other. The model generalizes the Frenkel-Kontorova model, replacing the rigid potential by a second deformable chain.

2. Double Chain Model

Figure 1 visualizes the Double Chain Model. The intrachain interaction is harmonic, the interchain interaction between the chains is Lennard-Jones like. In previous work[4] a Gaussian potential was used as the interchain potential. The potential energy of the model is written as:

$$E = \tfrac{k_1}{2}\sum_i (x_i - x_{i-1} - \alpha)^2 + \tfrac{k_2}{2}\sum_j (y_j - y_{j-1} - \beta)^2 + 2k_{12}\sum_{i,j}\Phi(x_i - y_j) \quad (1)$$

where k_1 and k_2 are the force constants of the intrachain potentials, α and β the equilibrium distances, x_i and y_j the positions of the i-th (j-th) molecules of the first and second chain respectively. The masses of the molecules are taken unity. Note that the model is one-dimensional. The molecules are allowed to move only along the chain axes.

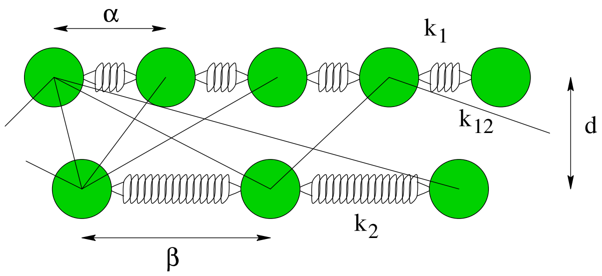

Figure 1: The Double Chain Model. The intrachain potentials are harmonic, the interchain potential Lennard-Jones like. The distance d is fixed, the periods in chain 1 and 2 are α and β respectively.

Numerical simulations use approximants to a real incommensurate system. They are made by considering a large unit cell containing p molecules in one chain and q molecules in the other chain. In the calculations, the ratio p/q approximates the golden mean.

The interchain potential was chosen to be a Lennard-Jones potential. This potential is long range, in contrast to the earlier used Gaussian potential that acts within one unit cell.

3. Phase diagram

The dispersion relations lead to two different cases. If the interchain coupling is weak compared to the intrachain couplings, two zero frequency modes in Γ are degenerated in the incommensurate case. One corresponds to the acoustic mode, the other to the phason or sliding mode. In the latter mode the two chains move in opposite directions. As the interchain potential increases, the degeneracy breaks, and a phason gap appears. This gap is defined as the difference between the frequencies of the phason and acoustic mode.

The breaking of degeneracy is a phase transition from an analytic to a nonanalytic region. The force constants determine in which region the system is. However, one of them, k_{12}, acts as a scaling parameter, whereas the other two determine the shape of the transition line. Figure 2 shows the phase diagram of an approximant, which has 55 molecules in one chain and 34 in the other. In the case of the Lennard-Jones potential the transition line is asymmetric and is not convex. It turned out that other ratios p/q, with $47 \le p \le 59$ and $q=34$, gave more and less symmetric diagrams[6]. The commensurate ratio 51/34 did not show a transition line. Consequently, the approximant is not a special case, i.e. it is not an extremum beyond which no other transition lines for other ratios are found.

In previous work[4] a Gaussian potential was used instead of the Lennard-Jones potential. Figure 2 shows the phase diagram in this case. The transition line is symmetric and does not have the bump. However, this Gaussian and the Lennard-Jones like potential differ much in range and strength. Therefore, another Gaussian was used, which has a well as deep as that of the Lennard-Jones

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