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Microscopic model for incommensurate crystal phases

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As a model for a crystal with an incommensurate phase we studied a linear chain of classical particles interacting by harmonic forces between up to third neighbors and anharmonic nearest-neighbor forces. Such a system has an infinite number of stable equilibrium configurations, among which there are incommensurate ones. The system has translational invariance and consequently acoustic phonons. Other excitations may be identified as phasons and amplitudons. In a mean-field approximation the model exhibits a soft mode, a transition to an (in general) incommensurate phase, which develops via discommensurations to a superstructure at a lock-in transition.

I. INTRODUCTION

In the last few years one has investigated, both theoretically and experimentally, the properties of so-called incommensurate crystal phases. These phases are characterized by the simultaneous occurrence of two or more periodicities, which are incommensurate with respect to each other. As a consequence these systems do not have space-group symmetry in three dimensions. Nevertheless, they are by no means aperiodic or amorphous: They exhibit a perfect ordering with a great coherence length. The first examples were found in magnetic systems where the period of the magnetic helical ordering is incommensurate with the lattice periodicity.[1] Incommensurability was also discovered in ionic crystals, where the equilibrium positions show periodic displacements from a structure with lattice periodicity (displacive modulation),[2] or where the probability of finding an ion in one of several positions or orientations is given by an incommensurate periodic function (occupation modulation).[3] Other examples are quasi-one- or two-dimensional conductors or layered compounds, where the coupling between electrons and lattice is at the origin of a displacive modulation (charge-density-wave system),[4] intercalates, and other composite structures with two or more mutually incommensurate subsystems.[5,6] Also, layers of adsorbed atoms on a crystal surface may show an incommensurate structure.[7]

The origin of the incommensurate phase can be understood in case of charge-density-wave systems. It arises from an instability of the lattice at a wave vector twice the Fermi wave vector. For composite structures (and intercalates or adsorbed layers) the incommensurability is a consequence of the frustration produced by the various periodicities present. These systems have been studied extensively on one-dimensional theoretical models.[8–10] For ionic crystals the origin is less clear. Lattice-dynamical studies have shown that for certain values of the parameters a mode may become soft. If the wave vector of the soft mode is not on the Brillouin-zone boundary, this may give rise to an incommensurate phase.[11] The instability may be induced by a polarization catastrophe[12] if large polarizable ions are present. However, these studies cannot give information on what happens below the phase transition. There are phenomenological models based on a Landau theory[13,14] that have served to fix the ideas. A microscopic theory, however, is still lacking.

The incommensurate phase occurs as an intermediate phase between two ordinary crystal structures. This picture that arises from experiments and from the theories mentioned above is as follows. In the high-temperature phase the structure has space-group symmetry. In this phase a soft mode develops with decreasing temperature and leads to an instability of the crystal structure. Below T_i the crystal is modulated, sinusoidally just

below T_i, but with higher harmonics for lower temperature. This leads to a structure where the crystal shows domains, where the modulation is nearly commensurate, and domain walls (discommensurations), such that the overall periodicity of the modulation is incommensurate. As T decreases the volume of the commensurate regions grows until at $T=T_c$ the whole crystal becomes commensurate and a superstructure of the high-temperature phase is formed. Since the discommensurations can be found, in a phenomenological theory, as solutions of a sine-Gordon equation they are sometimes called solitons. In the original literature on charge-density-wave systems[4] and in the Landau-type theories excitations are predicted which are described as oscillations of the phase of the modulation function with respect to the underlying lattice. These excitations are called phasons. In the literature it is suggested that there is always one phason with zero frequency: The motion of the modulation function with respect to the lattice. This mode is also called a sliding mode.

The phenomena of phasons and sliding modes have been studied in a microscopic model for composite structures, the Frenkel-Kontorova model. It studies a one-dimensional linear chain in a periodic background potential. Because this model does not have translational invariance it is not well adapted as model for ionic crystals. Other microscopic models giving rise, in principle at least, to incommensurate phases lack also this property.[15,16] Therefore, we want to study a translationally invariant model for an incommensurate phase. In such a model one can investigate, next to phonons, also possibly other excitations, like phasons, and the nature of discommensurations, if they occur as dynamical features. As such a model we consider a linear chain with short-range interaction (up to third-nearest neighbors) and anharmonic forces.

In Sec. II the model is introduced. The stationary configurations are discussed first for only nearest-neighbor and next-nearest-neighbor interactions in Sec. III. These configurations are treated, including third-nearest-neighbor interactions in Sec. IV. Not all stationary configurations correspond to minima of the potential energy. Their stability is discussed in Sec. V. For stable configurations the vibrations around the minima are studied in Sec. VI. These excitations influence the thermodynamic properties and are responsible for a renormalization of the coupling constants. In Sec. VII it is shown that in a mean-field approximation one can construct a phase diagram. The first sections are restricted to the case of negative next-nearest-neighbor interaction. For a positive one a different behavior is found which is treated in Sec. VIII. Some of the properties of the system may be studied in a continuum approximation. This is done in Appendix A.

II. THE MODEL

In the theory of magnetism it is known that an instability with wave vector different from zero and away from the Brillouin-zone boundary may occur as a consequence of competition between nearest-neighbor and next-nearest-neighbor interactions, the two coupling constants being of different sign. With the use of such interactions the phase diagram for some magnetic model systems has been studied in Ref. 17. There superstructures occurred with periods up to six. For similar model systems, solutions to the (nonlinear) equations were also found with larger, practically speaking incommensurate, periods.[18,19] In a model for displacive modulation with only harmonic forces such an instability would lead to disintegration of the crystal. Therefore, it is necessary to take also anharmonic terms into account. Moreover, a qualitatively different behavior is found if also interaction with third-nearest-neighbors is considered. The presence of this neighbor term is essential to find some of the properties observed in experiment. As a result we are led to consider as a model for a crystal with displacive modulation a linear chain of classical particles with equal mass interacting via harmonic forces with up to the third-nearest neighbors and via a fourth-order term with the nearest neighbors. The basis structure of the model is an equidistant array with lattice constant a. The positions of the particles are given by their displacements u_n from the positions na in the basic structure. The Hamiltonian for our model is then given by

$$H=\sum_n \left[\frac{p_n^2}{2m}+\frac{\alpha}{2}(u_n-u_{n-1})^2+\frac{\beta}{2}(u_n-u_{n-2})^2 + \frac{\delta}{2}(u_n-u_{n-3})^2+\frac{\gamma}{4}(u_n-u_{n-1})^4 \right] . \tag{2.1}$$

Let us consider the equilibrium positions which follow from $\partial V/\partial u_n=0$, where V is the potential energy. This yields the equation

$$\alpha(2u_n-u_{n-1}-u_{n+1})+\beta(2u_n-u_{n-2}-u_{n+2})+\delta(2u_n-u_{n-3}-u_{n+3})+\gamma(u_n-u_{n-1})^3+\gamma(u_n-u_{n+1})^3=0\ . \quad (2.2)$$

We first consider the case $\beta<0$. The coupling parameter γ is always positive to ensure stability. Hence, one can choose new units of length and energy such that

$$\beta=-1,\ \ \gamma=+1\ . \quad (2.3)$$

It is convenient to introduce the difference coordinate:

$$x_n=u_n-u_{n-1}\ . \quad (2.4)$$

In terms of these coordinates the potential energy is

$$V=\sum_n\left[\frac{\alpha}{2}x_n^2+\tfrac{1}{4}x_n^4-\tfrac{1}{2}(x_n+x_{n+1})^2+\frac{\delta}{2}(x_n+x_{n+1}+x_{n+2})^2\right]\ . \quad (2.5)$$

The equations for the equilibrium positions in these variables become

$$\alpha(x_n-x_{n+1})-(x_n+x_{n-1}-x_{n+1}-x_{n+2})+\delta(x_n+x_{n-1}+x_{n-2}-x_{n+1}-x_{n+2}-x_{n+3})+x_n^3-x_{n+1}^3=0\ . \quad (2.6)$$

III. EQUILIBRIUM POSITIONS FOR $\delta=0$

First we consider the model with nearest- and next-nearest-neighbor coupling only: $\delta=0$. For this case a lower bound for V/L, where L is the number of particles, can easily be found. According to Eq. (2.4) we have

$$V=\sum_n\left[\frac{\alpha-2}{2}x_n^2-x_nx_{n+1}+\tfrac{1}{4}x_n^4\right]\geq\sum_n\left[\frac{\alpha-4}{2}x_n^2+\tfrac{1}{4}x_n^4\right]\ . \quad (3.1)$$

Hence for the potential energy per particle $\mathscr{V}$ we get

$$\mathscr{V}=V/L\geq 0 \ \text{ if } \alpha\geq 4$$
$$\mathscr{V}>\min\left[\frac{\alpha-2}{2}x^2+\tfrac{1}{4}x^4\right]=-\frac{(4-\alpha)^2}{4} \quad (3.2)$$
$$\text{if } \alpha\leq 4\ .$$

An obvious solution of Eq. (2.2) with $\delta=0$ is the basic equidistant structure

$$u_n=0,\ \ \mathscr{V}=0\ . \quad (3.3)$$

Another solution is the dilated (also equidistant) structure

$$u_n=ln,\ \ l^2=4-\alpha,\ \ \mathscr{V}=-(4-\alpha)^2/4\ . \quad (3.4)$$

Since these solutions reach the lower bound for $\mathscr{V}$ they describe the ground state, for $\alpha\geq 4$ and for $\alpha\leq 4$, respectively.

Equation (2.2) has also other solutions. To discuss these we rewrite the potential energy as

$$V=\sum_n\left[\frac{\alpha-4}{2}x_n^2+\tfrac{1}{4}x_n^4+\tfrac{1}{2}(x_n-x_{n+1})^2\right]\ . \quad (3.5)$$

For $\alpha<4$ this is the potential energy for a chain of particles in double-well potentials and connected by harmonic springs. Such a system has been studied by Aubry.[20] The dynamics of this not translational invariant system, however, are quite different from our system. This can already be seen from the presence of an acoustic phonon mode in the present system. For $4-\alpha>>1$ the wells are deep with respect to the harmonic coupling. Hence the particles will be near the bottoms of the wells:

$$|x_n|\simeq\sqrt{4-\alpha}\ . \quad (3.6)$$

The distribution over the two wells will not be random, because the harmonic coupling favors equal signs for x_n. If they are all the same one gets solution (3.4). Solutions with higher energy will also occur: A change of sign between adjacent particles will increase V. Therefore if other solutions exist, those with long intervals of x_n's with the same sign will be energetically lower.

For $0<4-\alpha<<1$ one can consider a continuum model. In this continuum approximation one can write for the potential energy

$$V=\int dx/a[(\alpha-4)\xi^2/2+\xi^4/4+a^2\xi'^2/2]\ . \quad (3.7)$$

The equation for the equilibrium positions in this approximation is

$$\rho\xi''=\sigma\xi+\xi^3 \quad (3.8)$$

with $\xi(na)=x_n$, $\rho=a^2$, and $\sigma=\alpha-4$. For $\sigma<0$ Eq. (3.8) has the solution

$$\xi(x)=A(k,\alpha)\mathrm{sn}(B(k,\alpha)x) \quad (3.9)$$

with $0 \leq k \leq 1$ the parameter of the Jacobi elliptic function sn and

$$A(k,\alpha)=k\left[\frac{8-2\alpha}{1+k^2}\right]^{1/2}, \quad B(k,\alpha)=\left[\frac{4-\alpha}{1+k^2}\right]^{1/2}. \tag{3.10}$$

The solution

$$x_n = A(k,\alpha)\,\mathrm{sn}(naB(k,\alpha)) \tag{3.11}$$

is a periodic function: The period of $\mathrm{sn}(k=0,x)$ is 2π, that of $\mathrm{sn}(k\to 1,x)$ goes to infinity. The energy of the solution and its dependence on k are discussed in Appendix A.

Other periodic solutions ($u_{n+N}=u_n$) of Eq. (2.2) can be found analytically.

$$N=2: \quad u_n=\tfrac{1}{2}(-1)^n\sqrt{-\alpha} \quad (\alpha<0)\,. \tag{3.12a}$$

$$N=3: \quad u_n=\tfrac{2}{3}\sqrt{1-\alpha}\cos\left[\frac{2\pi n}{3}+\varphi\right] \quad (\alpha<1)\,. \tag{3.12b}$$

$$N=4: \quad u_n=\sqrt{4-2\alpha}\sin\left[\frac{\pi n}{2}+\frac{\pi}{4}\right] \quad (\alpha<2)\,. \tag{3.12c}$$

The general case of solutions with an arbitrary period N can be obtained by solving numerically the set of $N-1$ nonlinear equations resulting from Eq. (2.5) when $x_{n+N}=x_n$. In view of Eq. (2.4) they satisfy the condition $\sum x_n=0$. For any N a periodic solution is actually found in this way. As an example in Fig. 1 solutions with period $N=20$ are given for a number of values of the parameter α. Plotted in the figure are the values x_n. For $4-\alpha<<1$ the function x_n is smooth and approximately sinusoidal. It corresponds to solution (3.11) of the continuum approximation with small k. For decreasing α the function becomes more edgy. The value of x_n are then given, approximately, by Eq. (3.6): The positions of the particles correspond to the bottoms of the wells of V.

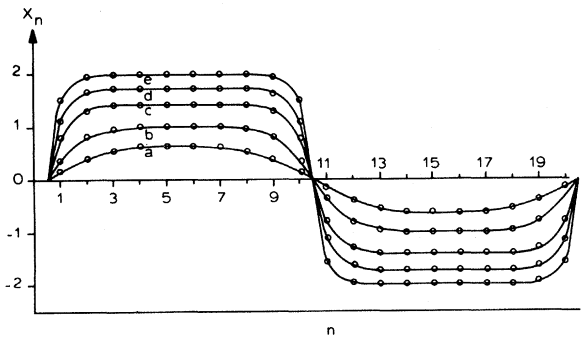

FIG. 1. Modulation function x_n for a periodic solution of Eq. (2.5) with period $N=20$, for various values of α. $\alpha=$ (a) 3.58, (b) 3.0, (c) 2.0, (d) 1.0, (e) 0.0.

The potential energy per particle of the solutions (3.12) is given by

$$\begin{aligned} &N=2: \quad \mathscr{V}=-\alpha^2/4\,, \\ &N=3: \quad \mathscr{V}=-(1-\alpha)^2/6\,, \\ &N=4: \quad \mathscr{V}=-(2-\alpha)^2/4\,. \end{aligned} \tag{3.13}$$

For higher N the value of $\mathscr{V}$ decreases towards the limit $-(4-\alpha)^2/4$ corresponding to $N\to\infty$.

Next to periodic solutions with one node per period there are also solutions with $2s$ ($s>1$) nodes per period. These solutions may be characterized by the sequence of numbers of positive and negative values of x_n. If one finds after the ith node p_i positive values followed by n_i negative ones, it is clear that

$$\sum_{i=1}^{s}(p_i+n_i)=N\,. \tag{3.14}$$

We may characterize such a configuration by

$$\langle p_1 n_1 p_2 n_2 \cdots p_s n_s \rangle\,.$$

A sequence of m identical numbers z may be abbreviated as z^m, a sequence of m identical pairs by $(p,n)^m$. The simple periodic solutions discussed before ($s=1$) correspond to the configurations $\langle N/2,N/2\rangle$ for even N and $\langle (N+1)/2, (N-1)/2\rangle$ for odd N. Since the sum of x_n is equal to zero, the particles cannot all be near the bottoms of the wells if N is odd. This explains why odd integer periods have a higher energy then even period ones.

The potential energy per particle decreases smoothly towards its minimum for increasing integer periods ($s=1$). The same is true if one considers $\mathscr{V}$ as a functions of N/s for series of configurations, for instance a series $\langle 3,4\rangle$, $\langle 3,3,3,4\rangle$, $\langle 3,3,3,3,3,4\rangle$, etc. The value of $\mathscr{V}$ for this series goes smoothly to its value for $N=6$, $s=1$. If one chooses for each N/s the minimal value of $\mathscr{V}$, it turns out that this is a smooth function of N/s. In Fig. 2 the value of $\mathscr{V}$ is plotted against the in-

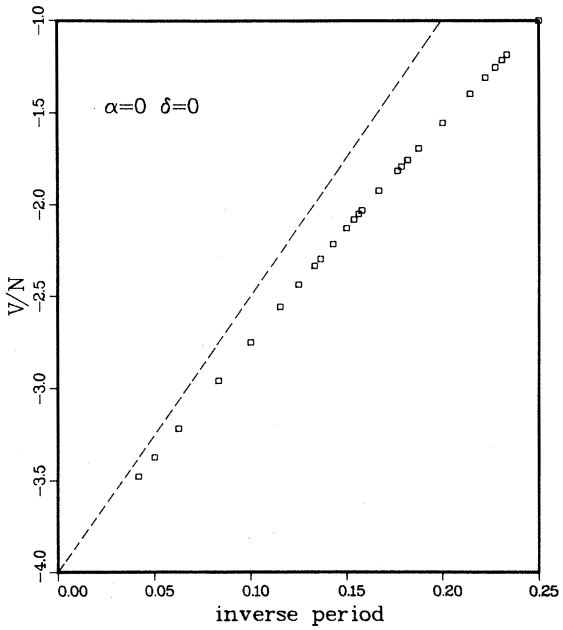

FIG. 2. Potential energy per particle against inverse fractional period s/N. The smooth curve is the energy density in the continuum approximation.

verse s/N for a number of sequences for $\alpha=0$. For this value of α the potential energy per particle is nearly a linear function of s/N. The total energy per period N is linear in s, i.e., linear in the number of kinks ($2s$) in the solution. Hence the energy per kink is almost constant. For $\alpha=0$ the kink energy is 6. For increasing α this energy decreases: $E_{\text{kink}} \simeq 6-2\alpha$ for $\alpha \lesssim 2$. At the same time the energy required for shifting the solution over one lattice constant goes to zero: the smooth solutions for $\alpha \simeq 4$ are weakly pinned.

The structure factor of a configuration may be defined as

$$F(q)=\frac{1}{N}\sum_{n=1}^{N}\exp[iq(na+u_n)]\,. \tag{3.15}$$

For a solution with period N and $2s$ nodes per period the main contribution to the structure factor is given by $q=\text{integer}\times 2\pi/a$. The next important values are reached for q values which differ from the former ones by a multiple of $(s/N)\times 2\pi/a$. The latter q values can be called satellites, the former main reflections. Then $|F(q)|^2$ shows the typical behavior of the x-ray pattern of a modulated crystal with modulation period N/s. In Fig. 3 a typical example is given. Hence the solution under discussion can be interpreted as modulated structures with an effective fractional period N/s. Because a number of properties, like $\mathscr{V}$ as seen before, depends smoothly on this period N/s it is possible to interpret solutions with large N but finite N/s as "nearly incommensurate." In this way one can say that in the present model there are also incommensurate solutions.

The structure of fractional period solutions is again smooth as long as $4-\alpha \ll 1$. For decreasing α the solutions are less regular. For $4-\alpha \gg 1$ the values of x_n will tend again to the value of (3.6). This implies that longer and shorter periods

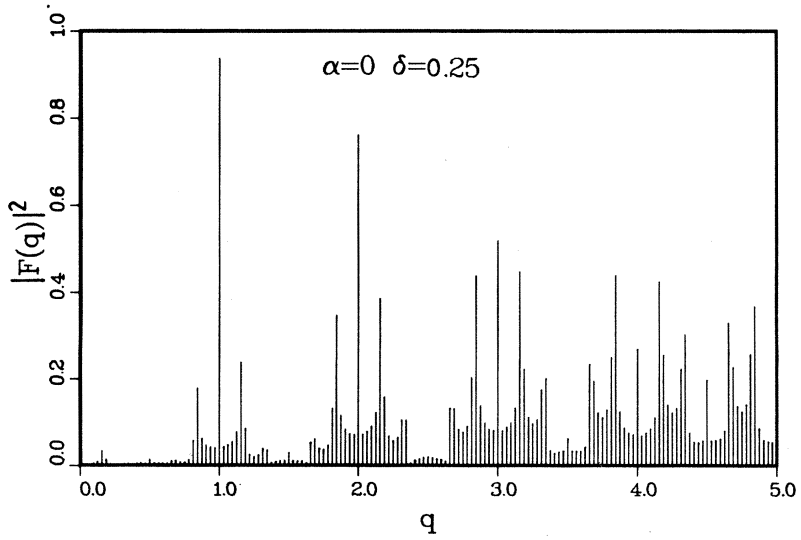

FIG. 3. Structure factor $|F(q)|^2$ corresponding to the solution of (2.5) plotted in Fig. 4.

alternate. A typical example is given in Fig. 4, where a solution is plotted with $N/s=6.4$. The solution has period 32 and 5 nodes per period. The configuration shows for part of the period nearly a period 6. After 4 of these quasiperiods an oscillation with period 8 brings the fractional period up to 6.4. This part of the solution may be interpreted as a discommensuration. These discommensurations occur if $4-\alpha>1$. For given N and s there may occur more than one solution. For example, next to the solution $\langle 3^8,4^2\rangle$ discussed above there is a solution $\langle 2^8,8^2\rangle$. Which of these solutions has lower energy depends on δ. For $\delta=0$ the solution $\langle 2^8,8^2\rangle$ has lower energy, for $\delta=0.25$ the configuration $\langle 3^8,4^2\rangle$, shown in Fig. 4, has this property. The reason is that in both cases the lower energy solution is more like the solution with absolutely minimal energy: the dilated solution for $\delta=0$, a period $N=6$ for $\delta=0.25$.

IV. EQUILIBRIUM POSITIONS FOR $\delta\neq 0$

If one considers also third-nearest neighbor interactions it follows from Eq. (2.4) that solutions with small values of $x_n+x_{n+1}+x_{n+2}$ are energetically favored. The trivial solution still exists: $u_n=0$ and $\mathscr{V}=0$. Also, a dilated equidistant solution exists if $\alpha<4-9\delta$. It is given by

$$u_n=ln,\quad l^2=4-\alpha-9\delta,\qquad \mathscr{V}=-(4-\alpha-9\delta)^2/4\ . \tag{4.1}$$

However, since the potential energy can now be written as

$$V=\sum_n\left[\frac{\alpha-4+9\delta}{2}x_n^2+\tfrac{1}{4}x_n^4+\frac{1-2\delta}{2}(x_n-x_{n-1})^2-\frac{\delta}{2}(x_n-x_{n-2})^2\right], \tag{4.2}$$

the solution (4.1) is not the one with minimal energy if δ is large enough.

Again there are solutions with integer and with fractional period. The potential energy per particle is a smooth function of N/s (Fig. 5). Increasing the parameter δ will increase the value of $\mathscr{V}$ for the solution (4.1) more than that with smaller periods. Hence if δ exceeds a certain value the curve $\mathscr{V}$ vs s/N will show a minimum at finite fractional period. This can also be seen from a continuum approximation as discussed in Appendix A. It implies that in order to have an incommensurate ground state it will be necessary to have interaction with third-nearest neighbors. On the other hand, this model can show such a minimum already for short-range interaction. Long-range interaction is not required to have an incommensurate structure.

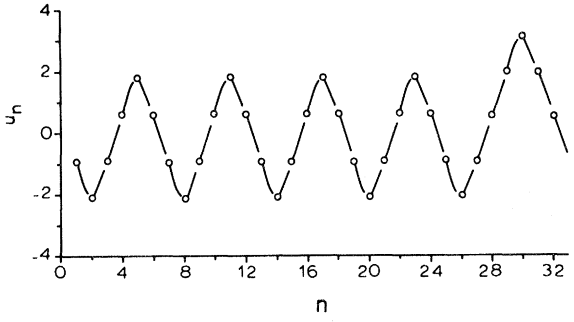

FIG. 4. Solution of Eq. (2.5) for fractional period $N/s=6.4$. $\alpha=0$, $\delta=0.25$.

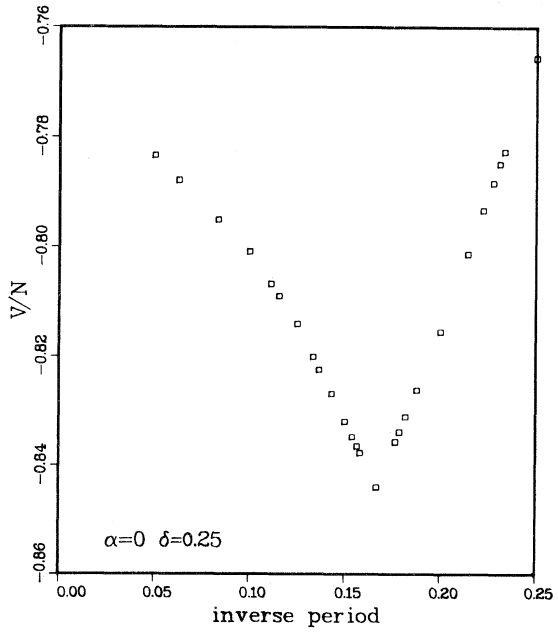

FIG. 5. Potential energy per particle against inverse fractional periods s/N for $\alpha=0$, $\delta=0.25$.

V. STABILITY

The solution given by Eq. (3.4) corresponds clearly to a stable minimum of the potential energy because it reaches its lower bound. The stability of other solutions can be checked by considering small displacements from the solutions and calculating the frequencies of the normal modes. If one denotes by ϵ_n the displacements from the positions u_n^0, which are solutions of the equilibrium equations, the positions are

$$u_n = u_n^0 + \epsilon_n \ . \tag{5.1}$$

The frequencies are the eigenvalues of the matrix

$$\left. \frac{\partial^2 V}{\partial \epsilon_n \partial \epsilon_m} \right|_{\epsilon=0} . \tag{5.2}$$

Hence the equations for the eigenvibrations are

$$\begin{aligned} m\omega^2 \epsilon_n = {} & (2\alpha - 2 + 2\delta + p_n + p_{n+1})\epsilon_n \\ & - (\alpha + p_{n+1})\epsilon_{n+1} - (\alpha + p_n)\epsilon_{n-1} \\ & + \epsilon_{n-2} + \epsilon_{n+2} - \delta\epsilon_{n-3} - \delta\epsilon_{n+3} \ , \end{aligned} \tag{5.3}$$

where

$$p_n = 3(u_n^0 - u_{n-1}^0)^2 \ . \tag{5.4}$$

The solution is stable if all the eigenvalues ω^2 of Eq. (5.3) are nonnegative.

For the trivial solution ($u_n = 0$) the eigenvalue equation becomes

$$m\omega^2 = 4\alpha \sin^2 \frac{q}{2} - 4 \sin^2 q + 4\delta \sin^2 \frac{3q}{2} \ . \tag{5.5}$$

The eigenvalues are nonnegative if

$$\alpha \geq 4 - 9\delta \quad \text{and} \quad 0 \leq \delta \leq \tfrac{1}{6}$$

or

$$\alpha \geq 1 + \frac{1}{4\delta} \quad \text{and} \quad \delta \geq \tfrac{1}{6} \ . \tag{5.6}$$

If the inequality signs apply there is only one mode with zero frequency: the rigid displacement of the whole crystal.

The eigenvalue equation for the dilated solution ($u_n = ln$) is given by

$$m\omega^2 = 8 \sin^2 \frac{q}{2} \left[-\alpha + 4 - 9\delta + 4 \sin^2 \frac{q}{2} + 4\delta \left[2 \sin^4 \frac{q}{2} - 3 \sin^2 \frac{q}{2} \right] \right] . \tag{5.7}$$

As a result the eigenvalues are nonnegative if

$$\alpha \leq 4 - 9\delta \quad \text{and} \quad 0 \leq \delta \leq \tfrac{1}{6}$$

or

$$\alpha \leq 4 - 9\delta - \frac{(1-6\delta)^2}{8\delta} \quad \text{and} \quad \delta \geq \tfrac{1}{6} \ . \tag{5.8}$$

For the solutions with simple periods $N = 2,3,4$ the stability conditions can be calculated analytically and are given in Appendix B. The stability for the other solutions follows from a numerical calculation. For each solution a region in the $\alpha\delta$ plane can be given in which the solution is stable. A number of stability limits are plotted in Fig. 6. From this we see that if δ increases the dilated solution becomes unstable and a solution with finite period must have lower energy. For $\delta = 0$ the dilated solution has minimal energy. For this δ the periodic solutions are only stable for $N > N_0(\alpha)$, where $N_0(\alpha = 0) = 4$. For increasing α the value of $N_0(\alpha)$ increases.

VI. EXCITATIONS

The study of the stability of the solutions in the preceding section has given information on the stability regions. In this section we want to discuss the spectrum and the eigenvectors of the eigenvibrations around a stable solution.

Characteristic for our model is the existence of an $\omega = 0$ mode for every α and δ. It is a consequence of the translation invariance and corre-

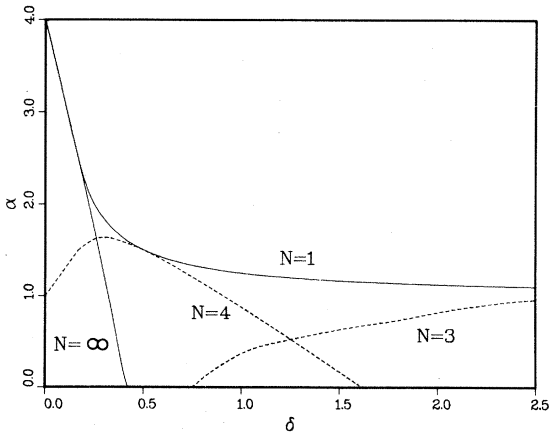

FIG. 6. Stability limits in the $\alpha\delta$ plane for some solutions of Eqs. (2.5). $N = 1$ is the basic structure, $N = \infty$ the dilated structure.

sponds to the acoustic phonon.

Inside its stable region the trivial solution has a spectrum determined by Eq. (5.5). Since the unit cell has only one particle there is no gap in the spectrum. Nevertheless, the dispersion curve is not always monotonically increasing. For decreasing α a dip in the dispersion curve develops. From Eq. (5.5) we see that the minimum of the curve becomes zero at the stability limit for a wave vector q_c given by

$$\cos(q_c a)=\begin{cases}1, & 0\le\delta\le\frac{1}{6}\\ (1-2\delta)/4\delta, & \delta\ge\frac{1}{6}\end{cases}. \tag{6.1}$$

An example of a changing dispersion curve is given in Fig. 7. The spectrum always runs from $\omega^2=0$ to $\omega^2=4\,(\alpha+\delta)/m$. For small q the modes correspond to sound waves. The sound velocity is given by $c=(\partial\omega/\partial q)_{q=0}$ so that

$$c=[(\alpha-4+9\delta)/m]^{1/2},\quad \alpha>4-9\delta\ . \tag{6.2}$$

For the dilated solution there is no gap either. Here the minimum in the dispersion curve occurs for q_c also given by Eq. (6.1) in the stability region as determined by Eq. (5.8). The spectrum runs from $\omega^2=0$ to $\omega^2=8(6-\alpha-13\delta)/m$. The sound velocity in such a structure is

$$c=\left[\frac{4-\alpha-9\delta}{2m}\right]^{1/2}. \tag{6.3}$$

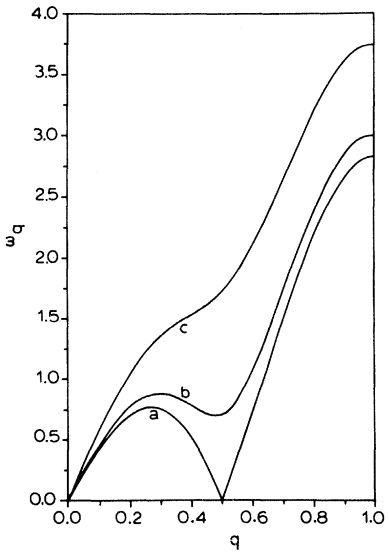

FIG. 7. Dispersion curve in the basic structure for varying value of α. $\alpha=$ (a) 1.5, (b) 1.75, (c) 3.0; $\delta=0.5$. For $\alpha=1.5$ the mode at $q_c=\pi/2a$ becomes soft.

As already mentioned in the preceding section for the finite period solutions with $N=3$ and 4 the spectrum can be found analytically (see Appendix B). In Fig. 8 the dispersion curves for these rather extreme cases are given for some values of α and δ. In the $N=4$ solution it is remarkable that, although the solution is a superstructure with four particles per unit cell, there is no gap in the spectrum. The spectrum of the $N=3$ solution has the peculiar property that, inside the stability region, there is always a double zero for $q=0$: One corresponds to the acoustic phonon, while the other is related to the freedom one has in φ in Eq. (3.12b). This is a mode which is known in the literature as a phason. Here it occurs, although the structure is commensurate. The other mode at vector $q=0$ corresponds to an oscillation in the amplitude. It is an amplitudon.

For other solutions the eigenvibration spectra can be calculated numerically. In Fig. 9 the spectra are plotted against the inverse fractional period s/N for a great number of solutions. In detail Fig. 9 is very complicated. There are, however, a number of global features which depend rather smoothly on the period. The most important one is the broad band of gaps connecting the lowest gaps for an even-integer period. Moreover, there are a number of structures which appear on different scales in different places in the figure. For α approaching 4 the structure becomes a bit more regular. The broad band shifts down and resembles the main gap in the spectrum of the modulated spring model.[21] The recursive structure found in that model cannot be seen here because here the amplitude of the modulation is not a free parameter.

If one considers the eigenvectors of the modes which correspond to the lower and upper boundary of the broad gap in Fig. 9, one sees that these modes describe oscillations in the phase and the amplitude, respectively, of the modulation function. In Fig. 10 an example is given for a solution together with the displacements in these two modes. In fact, these modes are the $2s$th and $(2s+1)$th mode at wave vector $q=0$ if one is not close to the stability limit. It should be noted that for $\alpha=0$, $\delta=0$ the frequency of the phason is not zero. Since the frequency is a smooth function of s/N this implies that also for incommensurate structures the phason frequency is not necessarily zero, contrary to what is suggested in the literature. Only near the stability limit does the frequency go to zero. As an example the phason frequency for the $N=4$ solution is given by

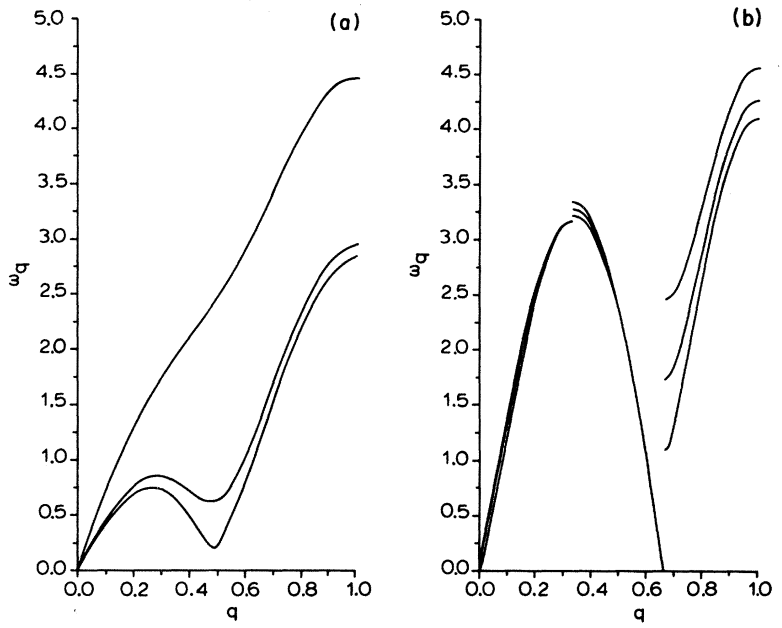

FIG. 8. Dispersion curves for the $N=3$ and $N=4$ solutions for a number of values of α. (a) $N=4$; $\delta=0.5$; $\alpha=0.0$, 1.41, 1.49. (b) $N=3$; $\delta=2.0$; $\alpha=0.0$, 0.75, 0.9.

$$m\omega^2_{\text{phason}}=4\left|\tfrac{3}{2}-\alpha\right| . \tag{6.4}$$

As will be discussed below there are indeed zero frequency phason modes if δ is sufficiently large.

The sound velocity is also one of the properties that depend smoothly on the period. The sound velocity versus fractional period is plotted in Fig. 11.

VII. THERMODYNAMIC PROPERTIES

The potential energy of our model has an infinite number of stable minima. To calculate the thermodynamic properties we only consider that part of the phase space that corresponds to harmonic vibrations around these minima. This amounts to the assumption that the local minima are separated by infinitely high walls. This is reasonable because the barriers between the minima of V are high, especially far from the stability limit of the trivial solution.

Because the potential wells are isolated from each other one can calculate the thermodynamic properties for each minimum separately. In this approximation one finds for the specific heat

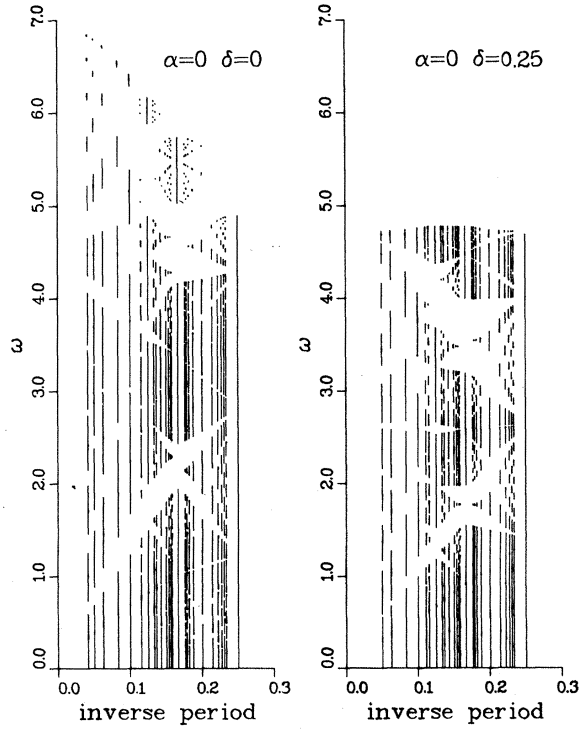

FIG. 9. Spectra of eigenvibrations around stable equilibrium positions versus the inverse fractional period. (For another choice of configurations see Ref. 27).

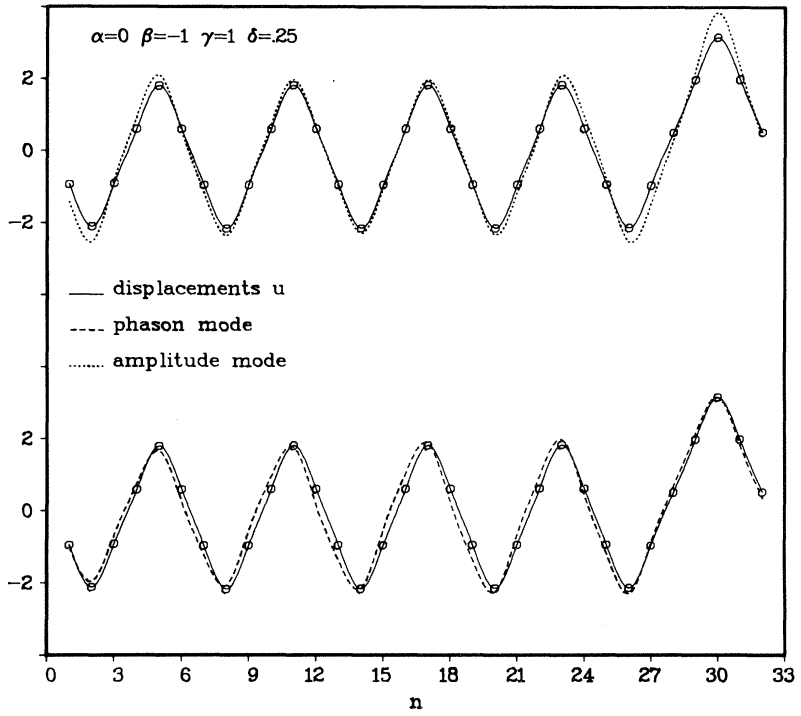

FIG. 10. Stable equilibrium positions with fractional period 6.4 together with the displacements in the phase and amplitude modes.

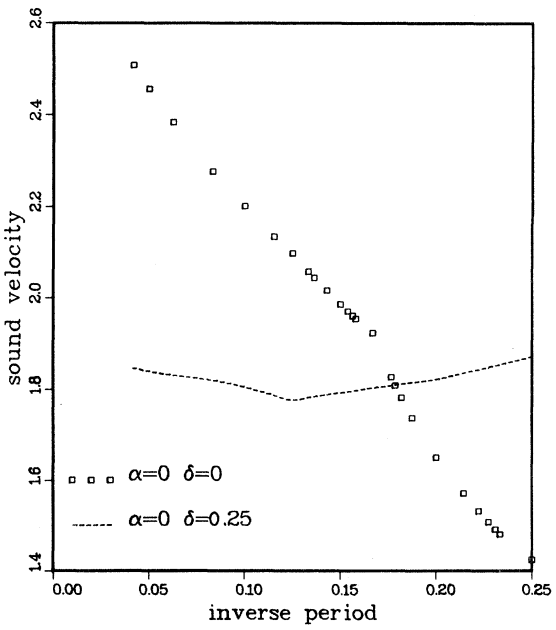

FIG. 11. Sound velocity vs inverse fractional period for $\alpha=0$, $\delta=0$ and $\alpha=0$, $\delta=0.25$.

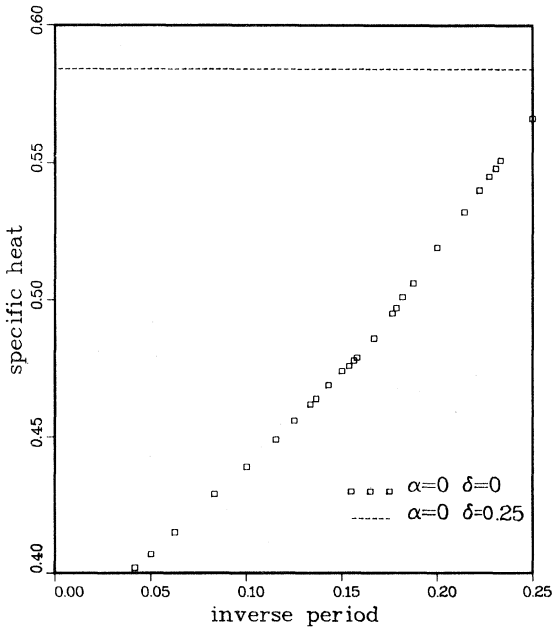

FIG. 12. Specific heat vs inverse fractional period for $\alpha=\delta=0$ and $\alpha=0$, $\delta=0.25$. For the latter case c_v is only approximately constant.

$$c_v=\frac{k_B\beta^2}{2\pi}\int_{-\pi}^{\pi}dq\sum_{j=1}^{N}\left[\frac{\omega_j(q)}{e^{\beta\omega_j(q)}-1}\right]^2 e^{\beta\omega_j(q)}, \tag{7.1}$$

where $\omega_j(q)$ are the N eigenvalues for Eq. (5.3) corresponding to wave vector q (N is the period of the solution). In Fig. 12 the specific heat is plotted for a number of (fractional) periods. It shows that c_v is a rather smooth function of the period.

It is well known that no phase transition can occur in a one-dimensional system with short-range interaction for $T\neq 0$. However, the model under consideration may describe well systems consisting of linear chains which are weakly coupled together while the interaction between the particles of each individual chain is described by Eq. (2.5). In order to get some idea about possible phase transition in such quasi-one-dimensional systems we may apply a simple mean-field approximation to our model. Taking the thermal average of the equations of motion for our model we get

$$m\langle\ddot{u}_n\rangle=-\alpha(2\langle u_n\rangle-\langle u_{n-1}\rangle-\langle u_{n+1}\rangle)+2\langle u_n\rangle-\langle u_{n-2}\rangle-\langle u_{n+2}\rangle-\delta(2\langle u_n\rangle-\langle u_{n-3}\rangle-\langle u_{n+3}\rangle)$$
$$-(\langle u_n-u_{n-1}\rangle^3-\langle u_{n+1}-u_n\rangle^3)+C_n-C_{n+1} \tag{7.2}$$

with

$$C_n=-[\langle(u_n-u_{n-1})^3\rangle-\langle u_n-u_{n-1}\rangle^3]\,. \tag{7.3}$$

We now make the approximation

$$\langle u_n^2u_m\rangle\simeq\langle u_n^2\rangle\langle u_m\rangle\,. \tag{7.4}$$

As a result Eq. (7.3) becomes

$$C_n\simeq-3[(\langle u_{n-1}^2\rangle-\langle u_{n-1}\rangle^2)\langle u_n\rangle-(\langle u_n^2\rangle-\langle u_n\rangle^2)\langle u_{n-1}\rangle]$$
$$-[(\langle u_n^2\rangle-\langle u_n\rangle^2)\langle u_n\rangle-(\langle u_{n-1}^2\rangle-\langle u_{n-1}\rangle^2)\langle u_{n-1}\rangle]\,. \tag{7.5}$$

Assuming that the thermal fluctuations in the displacements u_n do not depend on n, we find that

$$C_n\simeq-4(\langle u_n^2\rangle-\langle u_n\rangle^2)(\langle u_n\rangle-\langle u_{n-1}\rangle)\,. \tag{7.6}$$

Substituting Eq. (7.6) into Eq. (7.2) we see that the equations of motion for the thermal average of the displacements have the same form as for the displacements themselves with the only difference that there is a renormalization of the coefficient α. The nearest-neighbor effective coupling constant becomes α' with

$$\alpha'=\alpha+4(\langle u_n^2\rangle-\langle u_n\rangle^2)\,. \tag{7.7}$$

Hence a change in temperature will introduce a change in the effective parameter α'. Since the thermal fluctuations tend to increase with increasing temperature, a decrease in temperature corresponds in our model to moving downwards in the $\alpha\delta$ plane.

In Sec. V we have studied the stability limits of a number of solutions (Fig. 6). In the same $\alpha\delta$ plane one can investigate which solution corresponds to the lowest minimum for given α and δ. In Sec. III it has been shown that for $\delta=0$ and $\alpha<4$ this is the dilated solution (3.4). For δ sufficiently large the lowest-energy solution has a finite period. Numerically one finds that for $\alpha=0$, the dilated solution has the lowest energy if $\delta<0.24$. Between $\delta=0.24$ and 0.29 the simple period 6 has a lower energy, for δ between 0.29 and 0.90 another simple period ($N=4$) has this property. For $\delta>0.90$ the $N=3$ solution is stable, but there is always a solution with fractional period between 3 and 4 which has a lower energy. It is remarkable that simple periods (superstructures) play a dominant role. Only for bigger δ or for α, δ nearer to the stability limit of the trivial solution, there are cases with fractional (incommensurate) periods with lowest energy. In Fig. 13 the regions in the $\alpha\delta$ plane are indicated where the trivial, the dilated, and the simple period 4 and 6 solutions are the ones corresponding to the ground state. In the remaining regions there are still smaller domains where simple periodic solutions are the most stable (with, e.g., $N=8$), but in these regions occur also the "incommensurate" phases.

Summarizing the changes of the structure in this model as the temperature changes can be described as follows. For fixed temperature one moves along

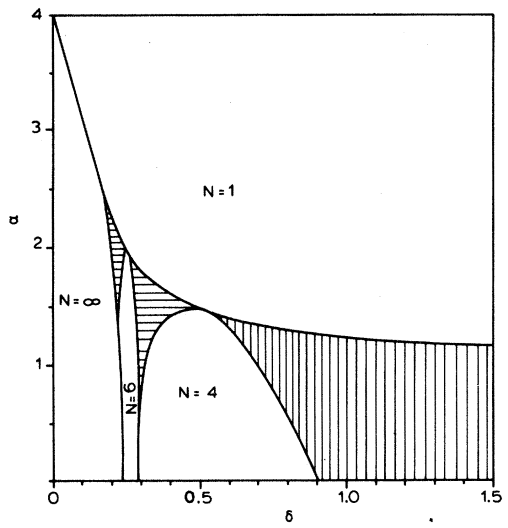

FIG. 13. "Phase diagram" indicating the structure of the ground state. In the vertically hatched region the ground state has a period between 3 and 4. In the horizontally hatched regions there are incommensurate ground states.

a vertical line in the $\alpha\delta$ plane. For high enough temperature the basic equidistant structure is stable. When T decreases a soft mode develops at wave vector q_c given by Eq. (6.1). At $T=T_i$ the frequency of this mode becomes zero and the basic structure becomes unstable. Below T_i the structure expands to a dilated, also equidistant, structure or to a sinusoidally modulated structure with wave vector q_c. In general, this is an incommensurate structure. When T decreases further the modulation runs through a number of fractional periods. For this temperature the modulation form is no longer sinusoidal, but has a sawtooth form. Some of the teeth correspond to a nearly simple period solution. These are alternating with teeth of different period which can be identified with "discommensurations." Just below T_i the soft mode has been split up into two modes: a phason and an amplitudon. The frequency of the phason is, in general, only nearly zero if one is near the stability limit. Finally, at still lower temperature there may be a transition to a simple periodic solution. It is a lock-in transition to a superstructure.

VIII. THE CASE OF POSITIVE NEXT-NEAREST-NEIGHBOR INTERACTION

In the magnetic one-dimensional models the structure is determined by the ratio of the nearest-neighbor coupling constant J_1 and the next-nearest-neighbor coupling J_2. The instability in the paramagnetic state occurst at wave vector q determined by $\cos(q)=-J_1/4J_2$. In our model this is not so: The local minima for $\alpha>0$, $\beta<0$ are different from those for $\alpha<0$, $\beta>0$. For positive second-nearest-neighbor coupling ($\beta>0$) the potential energy may be written after rescaling ($\beta=\gamma=+1$) as

$$V=\sum_n \left[\frac{\alpha}{2} x_n^2+\tfrac{1}{4}x_n^4+\tfrac{1}{2}(x_n+x_{n+1})^2 + \frac{\delta}{2}(x_n+x_{n+1}+x_{n+2})^2 \right] . \tag{8.1}$$

For large negative α the minima of V satisfy again Eq. (3.6), but now alternating signs are favored. This means that configurations with small (fractional) periods will, in general, have a lower energy. This picture may change if $|\delta|$ becomes large enough.

First we discuss some solutions of Eq. (2.2) and their stability. One of the equlibrium configurations is the trivial one:

$$x_n=0, \quad \mathscr{V}=0 . \tag{8.2}$$

The stability conditions follow from the eigenvalue equations of the oscillations around the solution

$$m\omega^2=4\sin^2(q/2)\{\alpha+4\cos^2(q/2) +\delta[4\cos^2(q/2)-1]^2\} . \tag{8.3}$$

The eigenvalues ω^2 are non-negative for arbitrary q if

$$\alpha>\begin{cases} -\delta & \text{if } -\frac{1}{2}\le\delta\le+\frac{1}{2} \\ -1+\dfrac{1}{4\delta} & \text{if } \delta\ge+\frac{1}{2} \\ -4-9\delta & \text{if } \delta\le-\frac{1}{2} \end{cases} . \tag{8.4}$$

When α approaches the stability limit the frequency of the mode with wave vector q_c goes to zero, where

$$\cos(q_c a)=\begin{cases} -1 & \text{if } -\frac{1}{2}\le\delta\le+\frac{1}{2} \\ -(1+2\delta)/4\delta & \text{if } \delta\ge+\frac{1}{2} \\ +1 & \text{if } \delta\le-\frac{1}{2} \end{cases} . \tag{8.5}$$

For values of α below the stability line the basic structure is certainly no longer the ground state. Other solutions (with small integer periods) are the following:

$$
\begin{aligned}
&N=2:\ x_n=(-1)^n\sqrt{-\alpha-\delta}, && \mathscr{V}=-(\alpha+\delta)^2/4 && (\alpha+\delta<0),\\
&N=3:\ x_n=2\sqrt{-\alpha-1}\cos\left[\frac{2\pi n}{3}+\varphi\right], && \mathscr{V}=-(1+\alpha)^2/6 && (\alpha<-1),\\
&N=4:\ x_n=x\sin\left[\frac{\pi n}{2}+\frac{\pi}{4}\right],\ x^2=-2(2+\alpha+\delta), && \mathscr{V}=-(2+\alpha+\delta)^2/4 && (\alpha+\delta+2<0).
\end{aligned}
\tag{8.6}
$$

The stability limits for these solutions may be calculated exactly (see Appendix B). They are plotted in Fig. 14. Also a dilated equidistant configuration may form a stable minimum:

$$
\begin{aligned}
&x_n=l,\ \ l^2=-(\alpha+4+9\delta),\\
&\mathscr{V}=-(\alpha+4+9\delta)^2/4 .
\end{aligned}
\tag{8.7}
$$

Its stability limit is also indicated in Fig. 14. A peculiar point in the diagram is the point $\alpha=\frac{1}{2}$, $\delta=-\frac{1}{2}$, where the solutions with $N=1$ (basic structure), $N=\infty$ (dilated structure), and $N=2$ all become unstable.

We did not investigate the phase diagram for $\beta=+1$ as extensively as that for $\beta=-1$. However, four main regions may be distinguished: In the upper right-hand corner the basic structure ($N=1$) is the ground state. In the region including the negative α axis lies the region where the antiferrodistortive structure ($N=2$) is the ground state. The boundary of this region is not indicated, only the stability limit of this solution. For sufficiently negative values of δ these two solutions become unstable for excitations with zero wave vector. To the left of this stability limit is the region where the dilated state is the ground state. Finally in the right-hand lower corner is the region where the ground state has a fractional period between 2 and 3. An example of a ground state for $\alpha=-1.5$ and $\delta=+1.5$ is given in Figs. 15 and 16 where both its modulation function u_n and its dispersion relation are given. It is noticeable that in this case there is again a double zero at wave vector zero corresponding to a displacement mode and a phason. So although one is away from the stability limit there is a phason with zero frequency. The reason is that the modulation function u_n is sinusoidal in this case (Fig. 15). The same happens for $\beta<0$ in the region in the $\alpha\delta$ plane where the ground state has a period between 3 and 4. For both $\beta<0$ and $\beta>0$ this zero frequency phason mode is inherited from that in the simple $N=3$ configuration, which is discussed in Appendix B.

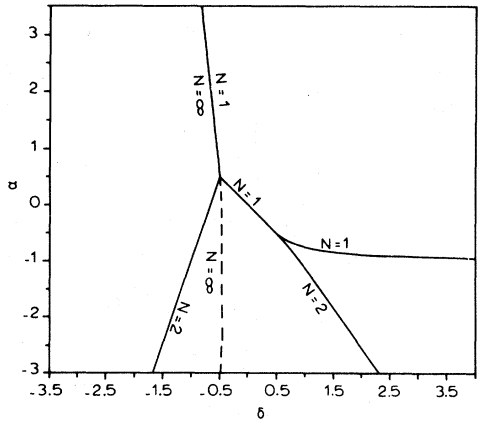

FIG. 14. Stability limits for some solutions of Eq. (2.2) with $\beta=+1$.

IX. DISCUSSION

On the example of a simple one-dimensional model it was shown that even for short-range interaction an incommensurate crystal phase may occur as ground state. The origin of this incommensurability is the competition between the interactions with the first-, second-, and third-nearest neighbors. The interaction with third-nearest neighbors is essential to have a finite modulation period. The model is applicable for ionic and molecular crystals because the incommensurate phase does not arise from interaction with another

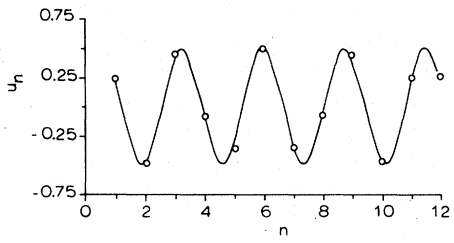

FIG. 15. Modulation function u_n for the ground state at $\alpha=-1.5$, $\beta=+1$, and $\delta=+1.5$. The curve is the function $A\cos[q(x-6a)]$ with $q=8\pi/11a$.

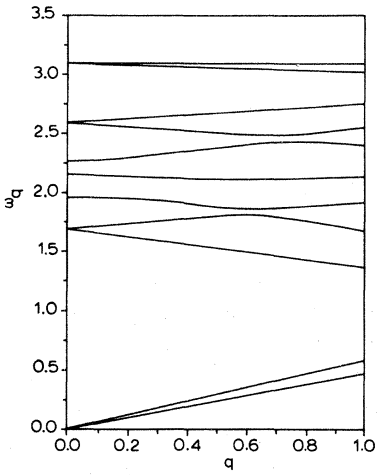

FIG. 16. Dispersion relation for the solution of Fig. 15. Notice the acoustic phonon and the phason branch which both go to zero for $q=0$.

(sub)system. Moreover, it is translationally invariant. For most values of the interaction parameters the ground state is the basic equidistant structure, a dilated equidistant structure with other lattice constant or a superstructure.

The model has an finite number of stable equilibrium configurations. In energy these are, in general, separated by high barriers. This will have consequences for the thermodynamical behavior: At not too high a temperature the system may stay for a long time in one of the configurations which is not the ground state. Relaxation to the ground state will then be slow. This is especially so outside the direct environment of the phase transition line, where the barriers are still relatively low. However, near the phase transition to the superstructure this slowing down can be important.

The excitation spectra show only a small number of gaps if the form of the modulation is simple. This is in agreement with the results for a one-dimensional model with modulated spring constants.[21] In general, however, the spectra are rather complex with many gaps. Nevertheless, there are structures and certain bands which change only slowly as the wave vector of the modulation is changed. The most important of these is a gap originating from the wave vector $2\pi s/Na$, where N is the actual period and $2s$ the number of nodes in one period.

In a simple mean-field approximation a change in temperature means a change in the interaction parameters. In this way the parameter space may be considered as a phase space. In this approximation one may have for decreasing temperature the following sequence of changes. First a soft mode develops: One of the phonon modes gets a zero frequency (of course this is well known) at temperature T_i. Below $T=T_i$ the structure has a sinusoidal modulation (in general incommensurate). For still lower T the modulation becomes more edgy and at $T=T_c$ a phase transition to a superstructure is possible. This sequence of events depends strongly on the parameters and the change of the parameters with temperature.

The model clarifies the nature of excitations which are characteristic for incommensurate phases: the phasons and amplitudons. These have been introduced in phenomenological theories, but it turns out that they appear also as excitations in a dynamical model. These excitations describe oscillations in the phase and the amplitude, respectively, but they are just phonons. The phonons belonging to the direct environment of such a point in the dispersion curve can then be said to belong to the "phason branch." The name "phason" only describes a main property of the mode. Actually the amplitude of the mode changes also. (In the case of a sinusoidal modulation it is just a phase mode.) Only near the transition temperature T_i or in the region in the phase diagram where the ground state has a period between 2 and 3 (if $\beta>0$), or between 3 and 4 (if $\beta<0$), the frequency of the phason is (nearly) equal to zero. In general it is not, even not for incommensurate phases. This can be an explanation why it is difficult to find the phason.[22,23] In the literature it is suggested that its frequency is always zero in an incommensurate phase. Notice, however, that a phason with zero frequency may even exist in a commensurate phase.

It is common to consider changes in the phase of the modulation function which are decoupled from the amplitude changes. In this way it is possible to write down a sine-Gordon equation for the phase.[24] The solutions are the discommensurations (or solitons) which one believes to have seen experimentally.[25,26] One should be careful with the interpretation, because, as our model shows, the discommensurations do not correspond to a mere phase change. In general the amplitude changes also (cf. Fig. 4). However, if one assigns a phase to each value of u_n (e.g., if A is the maximal value in a half-period by definition $\cos\varphi_n=u_n/A$) one may consider the difference between this phase and that of a simple periodic configuration. In Fig. 17

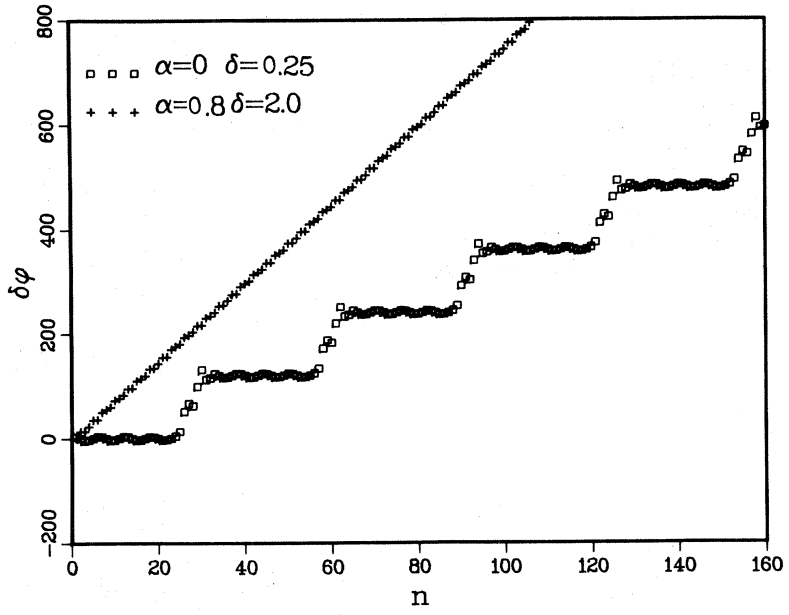

FIG. 17. The difference in phase between a solution and a regular sinusoidal function. (a) The solution of Fig. 4 compared with a period $N=6$ structure ($\alpha=0.0$, $\beta=-1$, $\delta=0.25$) (b) the ground state at $\alpha=0.8$, $\beta=-1$, $\delta=2.0$ compared with a sinusoidal function with period 3. In this case the solution itself is nearly sinusoidal, like the solution of Fig. 15.

this difference is plotted for the solution of Fig. 4 compared with a period $N=6$ configuration. The resulting graph is very similar to the one obtained from the sine-Gordon equation in a continuum approximation.[24]

The sound velocity and the specific heat are two closely related properties which depend smoothly on the modulation wave vector. For simple odd-integer periods both quantities have anomalies.[27] The results presented in Ref. 27 differ slightly from those presented here, because of another choice of sequences of solutions. In Ref. 27 we included also solutions of the type $\langle n^p,m\rangle$ whereas here we considered only sequences $\langle n^p,m^q\rangle$ with p and q even which correspond, in general, to solutions with lower energy. Moreover, in these sequences the anomalies at odd-integer periods do not occur so that the curves are smoother.

Although the model is only one dimensional one may assume that the main characteristics apply also to two- and three-dimensional systems. Actually, the use of a mean-field approximation for the study of phase transitions is only justified if we consider the model as an approximation for a higher-dimensional system with strong anisotropy. A preliminary calculation shows that many features carry over to higher-dimensional models.

APPENDIX A: CONTINUUM APPROXIMATION

In this appendix we present some results for our model in the continuum limit. Equation (3.8), which gave a first indication of the form of the solutions of Eq. (2.2), can be generalized for arbitrary values of the parameters α and β. To that end we write the potential energy V as

$$V=\sum_n[\alpha' x_n^2/2+\beta'(x_n-x_{n-1})^2/2 +\delta'(x_n-x_{n-2})^2/2+x_n^4/4]\,, \tag{A1}$$

where

$$\alpha'=\alpha+4\beta+9\delta,\quad \beta'=-\beta-2\delta,\quad \delta'=-\delta\,. \tag{A2}$$

In the continuum approximation one may write

$$aV=\int dx[\sigma\xi^2/2+\xi^4/4+\rho(\xi')^2/2] \tag{A3}$$

with

$$\sigma=\alpha+4\beta+9\delta,\quad \rho=-\beta-6\delta\,. \tag{A4}$$

The minimal energy is obtained for a solution of the differential equation

$$\rho\xi''=\sigma\xi+\xi^3 , \tag{A5}$$

which may be integrated yielding

$$x=x_0+\int_{\xi_0}^{\xi} dt[(t^4+2\sigma t^2+2c)/2\rho]^{-1/2} , \tag{A6}$$

where c is an integration constant. The solutions can be distinguished according to the sign of ρ (i.e., the sign of the leading polynomial coefficient) and the sign of the roots $-\sigma\pm(\sigma^2-2c)^{1/2}$ of this polynomial.

Case I: $\rho>0$, $\sigma<0$

For $0\le c\le\sigma^2/2$ both roots are positive: $a^2=-\sigma+(\sigma^2-2c)^{1/2}$ and $b^2=-\sigma-(\sigma^2-2c)^{1/2}$. The solution (A6) is in this case

$$x=\sqrt{2\rho}\int_0^{\xi}\frac{dt}{[(t^2-a^2)(t^2-b^2)]^{1/2}}$$

or

$$\xi=b\,\mathrm{sn}\left[x\frac{a}{\sqrt{2\rho}},\frac{b}{a}\right] , \tag{A7}$$

where $\mathrm{sn}(x,k)$ is a Jacobi elliptic function with modulus $k=b/a$.[28] For $c<0$ one of the roots is negative and the integral diverges.

Case II: $\rho<0$, $\sigma<0$

For $c<0$ the roots may be denoted by $a^2=-\sigma+(\sigma^2-2c)^{1/2}$ and $-b^2=-\sigma-(\sigma^2-2c)^{1/2}$. Equation (A6) becomes

$$x=\sqrt{-2\rho}\int_{\xi_0}^{\xi}\frac{dt}{[(a^2-t^2)(t^2+b^2)]^{1/2}}$$

or

$$\xi=a\,\mathrm{cn}\left[x\left[\frac{a^2+b^2}{-2\rho}\right]^{1/2},\left[\frac{a^2}{a^2+b^2}\right]^{1/2}\right] , \tag{A8}$$

where $\mathrm{cn}(x,k)$ is an elliptic function with modulus $k^2=a^2/(a^2+b^2)$ which satisfies $\frac{1}{2}<k^2\le 1$.

For $0<c\le\sigma^2/2$ the roots are $a^2=-\sigma+(\sigma^2-2c)^{1/2}$ and $b^2=-\sigma-(\sigma^2-2c)^{1/2}$. Then the solution is

$$x=\sqrt{-2\rho}\int_{\xi_0}^{\xi}\frac{dt}{[(a^2-t^2)(t^2-b^2)]^{1/2}}$$

or

$$\xi=a\,\mathrm{dn}\left[x\frac{a}{\sqrt{-2\rho}},\left[\frac{a^2-b^2}{a^2}\right]^{1/2}\right] . \tag{A9}$$

The modulus k of $\mathrm{dn}(x,k)$ satisfies $0\le k\le 1$.

Case III: $\rho<0$, $\sigma>0$

For $c<0$ the roots are $a^2=-\sigma+(\sigma^2-2c)^{1/2}$ and $-b^2=-\sigma-(\sigma^2-2c)^{1/2}$. Now

$$x=\sqrt{-2\rho}\int_{\xi_0}^{\xi}\frac{dt}{[(a^2-t^2)(b^2+t^2)]^{1/2}}$$

or

$$\xi=a\,\mathrm{cn}\left[x\left[\frac{a^2+b^2}{-2\rho}\right]^{1/2},\left[\frac{a^2}{a^2+b^2}\right]^{1/2}\right] , \tag{A10}$$

i.e., the same expression as Eq. (A8), but now $0\le k^2<\frac{1}{2}$. For $0\le c\le\sigma^2/2$ expression (A6) is not real.

Hence for all three cases there are periodic solutions. Notice, however, that only the sn and cn functions satisfy the condition that $\int_0^{4k}\xi(x)dx=0$, the dn function does not. So the latter does not correspond to a periodic solution in the displacements.

The potential energy density can be calculated using Eq. (A3):

$$\mathscr{V}=\frac{1}{K(k)}\int_0^{K(k)}dx[\sigma\xi^2/2+\xi^4/4+\rho(\xi')^2/2] , \tag{A11}$$

where $K(k)$ is one quarter of the period of the Jacobi elliptic functions. The resulting expressions are the following.

Case I: $\rho>0$, $\sigma<0$

We have

$$\mathscr{V}=-\frac{c}{6}+[\sigma^2-(\sigma^2-2c)^{1/2}]/3+\sigma[\sigma-(\sigma^2-2c)^{1/2}]\frac{E(k)}{3K(k)} , \tag{A12}$$

where $E(k)$ is the second complete integral. In this region, $\delta<-\beta/6$ and $\alpha<-4\beta-9\delta$, the solution is an elliptic sn function with modulus

$$k^2=\frac{-\sigma-(\sigma^2-2c)^{1/2}}{-\sigma+(\sigma^2-2c)^{1/2}} . \tag{A13}$$

For $c \simeq 0$ one has $k \simeq 0$. Hence the solution is a sine function with amplitude $\sqrt{-c/\sigma}$ and $\mathscr{V} \simeq 0$. For $c = \sigma^2/2$, one gets $k = 1$ and the solution is a tanh function with amplitude $\sqrt{-\sigma}$ and $\mathscr{V} = -\sigma^2/4$. The latter is the most stable solution (minimal energy). It is the dilated solution (3.4) with one kink.

Case II: $\rho < 0$, $\sigma < 0$

We have

$$\mathscr{V} = \frac{c}{4} + [\sigma + (\sigma^2 - 2c)^{1/2}] \times \left[-\frac{\sigma}{2} + \frac{1}{6}(\sigma^2 - 2c)^{1/2} \right] + \tfrac{2}{3}\sigma(\sigma^2 - 2c)^{1/2} \frac{E(k)}{K(k)} . \quad \text{(A14)}$$

In this region, $\delta > -\beta/6$ and $\alpha < -4\beta - 9\delta$, the solution with $c < 0$ is a cn function with modulus

$$k^2 = [-\sigma + (\sigma^2 - 2c)^{1/2}/2(\sigma^2 - 2c)^{1/2}] . \quad \text{(A15)}$$

For $c \simeq 0$ the modulus $k \simeq 1$. The solution then is a 1/cosh function with amplitude $\sqrt{-2\sigma}$ and $\mathscr{V} = 0$: It is a localized state. For $c \to -\infty$ the modulus k^2 tends to $\frac{1}{2}$ and $\mathscr{V}$ to $-c/12$, which goes to positive infinity. However, as a function of c the potential energy has a minimum at finite c, i.e., at finite k and finite period: $c = -1.5\sigma^2$, $k^2 = \frac{3}{4}$, period $\simeq 6\sqrt{\rho/\sigma}$, $\mathscr{V} \simeq -0.29\sigma^2$. The depth of the minimum in the continuum approximation is in agreement with the depth in the discrete calculation (Fig. 5). The predicted position of the minimum, however, is off by a factor. This is due to the fact that for the minimum one has inverse period 0.16, which cannot very well be described in a continuum approximation.

Solution (A9) with $0 < c < \sigma^2/2$ has a potential energy density

$$\mathscr{V} = \frac{c}{6} - 2\sigma(\sigma^2 - 2c)^{1/2} + [-\sigma + (\sigma^2 - 2c)^{1/2}] \frac{\sigma}{3} \frac{E(k)}{K(k)} . \quad \text{(A16)}$$

For $c = 0$ one has $k = 1$. Hence the solution is a 1/cosh function with $\mathscr{V} = 2\sigma^2$. For $c = \sigma^2/2$ one has $k = 0$. Then the solution is a constant $\sqrt{-\sigma}$. It corresponds to the dilated solution (3.4) and $\mathscr{V} = -\sigma^2/4$.

Case III: $\rho < 0$, $\sigma > 0$

We have

$$\mathscr{V} = \frac{c}{4} + [\sigma + (\sigma^2 - 2c)^{1/2}] \left[-\frac{\sigma}{2} + \frac{1}{6}(\sigma^2 - 2c)^{1/2} \right] + \tfrac{2}{3}\sigma(\sigma^2 - 2c)^{1/2} \frac{E(k)}{K(k)} . \quad \text{(A17)}$$

This is the same expression as (A14). Again $\mathscr{V} = 0$ for $c = 0$ and for $c \to -\infty$ the modulus k^2 tends to $\frac{1}{2}$ and $\mathscr{V}$ to $-c/12 > 0$. However, now $\mathscr{V}$ is positive for all c. Hence this solution has always higher energy than the null solution.

Because x_0 in (A6) is an integration constant, the solutions (A7)–(A10) represent a choice of this constant. Other solutions are obtained by a simple shift in x. Since this does not influence the energy density it means that in the continuum approximation there is always an excitation with zero frequency, corresponding to a shift of the modulation wave. This is the phason mode, which is not always present in the discrete case. In the continuum approximation it is the Goldstone mode corresponding to the translation symmetry of Eq. (A5).

APPENDIX B: ANALYTIC SOLUTIONS WITH PERIOD 2, 3, OR 4

For small-integer period N Eqs. (2.2) can be solved analytically. We give here the solutions and the parameter values of α and δ for which the solutions are stable.

For $N = 2$ one can write

$$x_n = (-1)^n A, \quad u_n = (-1)^n A/2 . \quad \text{(B1)}$$

The energy per particle then becomes

$$\mathscr{V} = (\alpha + \delta)A^2/2 + A^4/4 , \quad \text{(B2)}$$

which is a minimum for

$$A^2 = -\alpha - \delta \quad (\alpha + \delta \leq 0) . \quad \text{(B3)}$$

Because the value of p_n [Eq. (5.4)] is independent of n, the force constants are invariant under the lattice translation a. The frequency of the eigenmodes with wave vector q is given by

$$m\omega^2 = 4\sin^2\frac{q}{2} \left[-2\alpha - 3\delta + 4\beta\cos^2\frac{q}{2} + \delta\left[4\cos^2\frac{q}{2} - 1\right]^2 \right] . \quad \text{(B4)}$$

The solution is stable if $\omega^2>0$ for all q, i.e.,

$$\begin{aligned}
&\text{if } \beta=-1\text{: for } \delta\le\tfrac{1}{6},\ \alpha<-2+3\delta\\
&\qquad\text{for } \delta\le\tfrac{1}{6},\ \alpha<-(1+3\delta+1/4\delta)/2\\
&\text{if } \beta=+1\text{: for } \delta\le-\tfrac{1}{2},\ \alpha<2+3\delta\\
&\qquad\text{for } -\tfrac{1}{2}\le\delta\le\tfrac{1}{2},\ \alpha<-\delta\\
&\qquad\text{for } \delta\ge\tfrac{1}{2},\ \alpha<(1-3\delta-1/4\delta)/2\ .
\end{aligned}\tag{B5}$$

For $N=3$ the solution is of the form

$$x_n=A\cos(2\pi n/3+\varphi)\ .\tag{B6}$$

The potential energy per particle is

$$\mathscr{V}=(\alpha+\beta)A^2/4+3A^4/32\ ,\tag{B7}$$

which becomes minimal for

$$A^2=-4(\alpha+\beta)/3\quad(\alpha+\beta\le0)\tag{B8}$$

independent of the phase φ. Choosing $\varphi=0$ the solution is given by

$$u_n=A/2,0,-A/2\tag{B9}$$

for $n=1,2,3$ in the unit cell. The dynamical equation becomes

$$\mathrm{Det}\begin{vmatrix}
-3\alpha-3\beta+2\delta(1-\cos q)-m\omega^2 & \beta(1-e^{iq}) & (3\alpha+4\beta)e^{-iq}-\beta\\
\beta(1-e^{iq}) & 2\delta(1-\cos q)-m\omega^2 & \beta(1-e^{iq})\\
(3\alpha+4\beta)e^{iq}-\beta & \beta(1-e^{iq}) & -3\alpha-3\beta+2\delta(1-\cos q)-m\omega^2
\end{vmatrix}=0\ .\tag{B10}$$

For any α, β, and δ there are two modes with $\omega=0$ and $q=0$. For this value of q Eq. (B10) becomes

$$\mathrm{Det}\begin{vmatrix}
-3\alpha-3\beta-m\omega^2 & 0 & 3\alpha+3\beta\\
0 & -m\omega^2 & 0\\
3\alpha+3\beta & 0 & -3\alpha-3\beta-m\omega^2
\end{vmatrix}=0\ .\tag{B11}$$

One of the eigenvectors with $\omega=0$ is $(1,1,1)/\sqrt{3}$, which corresponds to an acoustic translation mode. The second one is $(1,-2,1)/\sqrt{6}$, which corresponds to a shift of the phase φ: It is the phason mode. The eigenvector for the mode with $m\omega^2=-6(\alpha+\beta)$ is $(1,0,-1)/\sqrt{2}$. It is an amplitude mode as one can see by comparing the eigenvector with Eq. (B9). The eigenvalues $m\omega^2$ of Eq. (B10) are positive for all q for sufficiently large δ.

For $N=4$ the solution is of the form

$$x_n=A\cos(\pi n/2+\varphi)\tag{B12}$$

for which the energy per particle is

$$\mathscr{V}=(\alpha+2\beta+\delta)A^2/4+(\sin^4\varphi+\cos^4\varphi)A^4/8\ .\tag{B13}$$

This is minimal for

$$A=-(\alpha+2\beta+\delta)\ \text{ if } \alpha+2\beta+\delta\le0\ .\tag{B14}$$

The corresponding solution in u_n is

$$u_n=0,A/2,0,-A/2\tag{B15}$$

for $n=1,2,3,4$ in the unit cell. Since the values of p_n [Eq. (5.4)] are independent of n, the force constants are invariant under a translation a. The eigenvalues of the normal modes are

$$m\omega^2=4\sin\frac{q}{2}\left[-2\alpha-6\beta-3\delta+4\beta\cos^2\frac{q}{2}+\delta\left[4\cos^2\frac{q}{2}-1\right]^2\right]\ .\tag{B16}$$

The solution (B15) is stable if $\omega^2>0$ for all q, i.e.,

$$\begin{aligned}
&\text{if } \beta=-1\text{: for } 0\le\delta\le\tfrac{1}{6},\ \alpha<1+3\delta\\
&\qquad\text{for } \delta\ge\tfrac{1}{6},\ \alpha<3-\delta-(1+2\delta)/8\delta\\
&\text{if } \beta=+1\text{: for } \delta\ge\tfrac{1}{2},\ \alpha<-5-3\delta+1/4\delta\\
&\qquad\text{for } -\tfrac{1}{2}\le\delta\le\tfrac{1}{2},\ \alpha<-3-\delta\\
&\qquad\text{for } \delta\le-\tfrac{1}{2},\ \alpha<-1+3\delta\ . \qquad \text{(B17)}
\end{aligned}$$

The eigenvalue (B14) for $q=\pm\pi/2$ corresponds to two modes which can be considered as degenerate phase and amplitude modes. The eigenvalue is

$$m\omega^2=-4(\alpha+2\beta+\delta)\ . \qquad \text{(B18)}$$

Notice that, contrary to the case $N=3$ there is no zero-frequency phase mode.

[1]A. Yoshimori, J. Phys. Soc. Jpn. 14, 807 (1959); A. Herpin, P. Meriel, and J. Villian, J. Phys. Radium 21, 67 (1960).

[2]G. C. Dubbeldam and P. M. de Wolff, Acta Crystallogr. B 25, 2665 (1969).

[3]H. Boehm, Habilitationsschrift, University of Munster, 1977 (unpublished); D. Kucharczyk, A. Pietrasko, and K. Lukaszewicz, Acta Crystallogr. A 34, S16 (1978).

[4]A. W. Overhauser, Phys. Rev. 128, 1437 (1962).

[5]C. K. Johnson and C. R. Watson, J. Chem. Phys. 64, 2271 (1976).

[6]J. P. Pouget, G. Shirane, J. M. Hasting, A. J. Heeger, N. D. Miro, and A. G. MacDiarmid, Phys. Rev. B 18, 3645 (1978).

[7]J. A. Venables and P. S. Schabes-Retchkiman, J. Phys. (Paris) 38, C4-105 (1977).

[8]J. Frenkel and T. Kontorova, J. Phys. (Moscow) 1, 137 (1939). F. C. Frank and J. H. van der Merwe, Proc. R Soc. London A Ser. 198, 205 (1949).

[9]S. Aubry, *Proceedings of the 8th International Colloquium on Group Theoretical Methods in Physics,* edited by L. P. Horowitz and Y. Ne'eman (Hilger, Bristol, 1980).

[10]J. E. Sacco and J. B. Sokoloff, Phys. Rev. B 18, 6549 (1978); A. D. Novaco, *ibid.* 22, 1645 (1980).

[11]M. S. Haque and J. R. Hardy, Phys. Rev. B 21, 245 (1980).

[12]C. M. Fortuin, Physica (Utrecht) 86A, 224 (1977).

[13]R. A. Cowley and A. D. Bruce, J. Phys. C 11, 3577 (1978); A. D. Bruce, R. A. Cowley, and A. F. Murray, *ibid.* 11, 3591 (1978); A. D. Bruce and R. A. Cowley, *ibid.* 11, 3609 (1978).

[14]Y. Ishibashi and V. Dvorak, J. Phys. Soc. Jpn. 44, 32 (1978).

[15]C. Haas, Solid State Commun. 26, 709 (1978).

[16]K. Michel Phys. Rev. B 24, 3998 (1981).

[17]T. Janssen, Phys. Kondens. Mater. 15, 142 (1972).

[18]P. Bak and J. von Boehm, Phys. Rev. B 21, 5297 (1980); P. Bak, Phys. Rev. Lett. 46, 790 (1981).

[19]M. E. Fisher and W. Selke, Phys. Rev. Lett. 44, 1502 (1980).

[20]S. Aubry, J. Chem. Phys. 62, 3217 (1975); 64, 3392 (1976); T. Schneider and E. Stoll, Phys. Rev. B 22, 5317 (1980).

[21]C. de Lange and T. Janssen, J. Phys. C (in press).

[22]H. Cailleau, F. Moussa, C. M. E. Zeyen, and J. Bouillot, Solid State Commun. 33, 407 (1980); A. H. Moudden, F. Denoyer, J. P Benoit, and W. Fitzgerald, *ibid.* 28, 575 (1978).

[23]J. D. Axe, M. Iizumi, and G. Shirane, Phys. Rev. B 22, 3408 (1980).

[24]P. Bak, in *Solitons in Condensed Matter Physics,* edited by A. R. Bishop and T. Schneider (Springer, Berlin, 1978).

[25]R. Blinc *et al.*, Solid State Commun. 34, 895 (1980); R. Blinc *et al.*, Phys. Rev. Lett. 21, 1406 (1981); V. Rutar *et al.* (unpublished).

[26]W. Rehwald and A. Vonlanthen, Solid State Commun. 38, 209 (1981).

[27]T. Janseen and J. A. Tjon, Phys. Rev. B 24, 2245 (1981).

[28]F. Oberhettinger and W. Magnus, *Anwendung der Elliptischen Funktionen in Physik und Technik* (Springer, Berlin, 1949).