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Time-dependent correlations in the exponential lattice. I. Formal theory of response functions

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Time-dependent response functions of classical lattice systems are the object of this study. A general representation for the displacement-displacement response function is presented. In the special case of a linear lattice with exponential interaction potential between adjacent particles this formula leads at low temperatures to a closed system of integrodifferential equations governing the time dependence of the response functions. Thus for this special type of lattice system which supports solutions with strong anharmonic features, a systematic study of dynamical correlations at low temperatures is possible. Numerical results will be presented in the accompanying paper.

I. INTRODUCTION

In recent years considerable attention has been devoted to the investigation of strong anharmonic phonon systems. Theories worked out within the context of conventional phonon theory completely fail to reproduce strong nonlinear features of solutions which describe the system in certain important regions of solution space.[1] Current research approaches the problem at the level of phenomenological models. Two important questions to be answered on the basis of the model assumptions concern the identification of elementary excitations of the system and the extent to which they may be considered independent within the frame of statistical mechanics. Krumhansl and Schrieffer[2] have provided—within the context of statistical mechanics—basic ideas which proved successful up to now. Since that time much effort has been devoted to the identification of fundamental modes of motion dominating the properties of the thermalized system, and much quantitative insight has been gained. Well-known examples are the sine-Gordon system[3,4] and the "ϕ^4" system.[5,6] In these models strong nonlinear localized modes are defined which are separated from the ground state by an energy gap. This characteristic facilitates the identification of these solitons within the static properties of the thermalized system.[4] In the case of the linear exponential lattice, however, the energy of the corresponding soliton modes continuously evolves out of the ground-state energy.[7] Identification seems more difficult in the static properties of the chain.

Nevertheless, first attempts to construct a configurational phenomenology have been undertaken.[8] Solitons in the exponential lattice reveal some similiarity with breather modes of the sine-Gordon system; both are characterized by a failure of an energy gap. It seems that breather solutions are most clearly identified in the dynamical properties of the sine-Gordon chain[9]: Recently it has been found[10,11] that not only solitons but also breathers give rise to specific excitation branches in the thermalized system. Results from inelastic neutron scattering experiments are available.[12] The observed central peak in the dynamic structure factor has been interpreted in terms of localized modes of the corresponding sine-Gordon system.[10,11,13]

This investigation is concerned with the dynamics of the exponential lattice. Dynamical correlation functions are important quantities: important with respect to their experimental significance and with respect to what they express physically. They reveal detailed information on the excitation dynamics of the system considered. Thus they provide an ideal reference point against which calculations based on models within the context of configurational phenomenology can be calibrated. Exact procedures for the computation of dynamical correlation functions do not exist, and approximate calculations based on some controllable analytic approximation procedure starting from first principles have not been worked out. Only molecular dynamic simulations are available, especially for the sine-Gordon[10,11] and the "ϕ^4" system.[14]

In this (I) and the following (II) paper a new approach is developed which provides microscopic calculations of dynamical correlation functions of classical lattice systems with exponential interaction potential between the constituent particles. This paper

will be concerned only with the formal procedure of this approach. The linear lattice with next-neighbor interaction is taken as an example to work out the approximations which allow a first-principles calculation of the response functions of relative displacements at low temperatures. The formalism described is along the lines with that one applied recently to the "ϕ^4" system (Bunde and Diederich[15]).

In Sec. II an exact expression for Kubo's response function is derived, representing it in terms of time-ordered products of correlations of the force constants corresponding to instantaneous particle positions. Up to this stage the calculation is valid for an arbitrary interaction potential.

In Sec. III this general formula is applied to an exponential lattice. Then the correlations of the force constants can be expressed by the correlations of the forces themselves, that is, by time derivatives of correlations of the particle positions. At low temperatures correlations of higher than second order may be neglected and rather involved time-ordered products can be disentangled. This approximation procedure leads us to a system of integrodifferential equations involving time-dependent memory functions. The memory kernels are expressed by the response functions themselves, thus resulting in a coupled system of nonlinear equations. In Sec. IV the conclusions are given.

In the following paper time-dependent relaxation functions of relative displacements between adjacent particles will be calculated numerically applying the formalism described to the linear exponential lattice. The dominant contributions to the corresponding spectral densities will be interpreted in terms of exact solutions of the equations of motion.

II. GENERAL THEORY OF THE DISPLACEMENT-DISPLACEMENT RESPONSE

With the present approach we are considering a one-dimensional lattice of classical particles with equal mass m and nearest-neighbor (NN) interaction potential $\phi(x_i - x_{i-1})$. x_i denotes the position of the particle at lattice site i (Cartesian coordinate). Following Toda[7] we introduce as the dynamical variables of the theory the distances between adjacent particles $R_i = x_i - x_{i-1}$. This leads to equations of motion

$$\begin{aligned} m\dot{R}_n &= 2S_n - S_{n+1} - S_{n-1} \ , \\ \dot{S}_n &= -\phi'(R_n) \ . \end{aligned} \tag{1}$$

Denoting the natural distance between two adjacent particles by D, Toda's exponential interaction potential may be written as

$$\phi(R) = (a/b)\exp[-b(R-D)] + a(R-D) \tag{2}$$

(a,b = const). For small values of $R - D$ we obtain the harmonic case with force constant ab.

Imposing periodic boundary conditions $S_1 = S_{M+1}$, $R_1 = R_{M+1}$, etc., we obtain a finite system of Eqs. (1) ($n = 1, \ldots, M$) which may be written in canonical form

$$\dot{R}_n = \frac{\partial H}{\partial S_n}, \quad \dot{S}_n = -\frac{\partial H}{\partial R_n}$$

with the Hamiltonian $H = K + W$:

$$K = \frac{1}{2m}\sum_{n=1}^{M}(S_n - S_{n+1})^2 \ , \tag{3}$$

$$W = \sum_{n=1}^{M}\phi(R_n) \ . \tag{4}$$

As the next step we introduce canonically conjugate variables A_k^1, A_{-k}^2 of wave-vector space [$-\pi < k = (2\pi/M)j \leq \pi$, j integer]

$$\begin{aligned} A_k^1 &= \frac{1}{\sqrt{M}}\sum_{n=1}^{M}\exp(-ikn)R_n \ , \\ A_k^2 &= \frac{1}{\sqrt{M}}\sum_{n=1}^{M}\exp(-ikn)S_n \ , \end{aligned} \tag{5}$$

and obtain equations of motion

$$\dot{A}_k^1 = \frac{\partial H}{\partial A_{-k}^2} = I_k A_k^2 \ , \tag{6}$$

$$\dot{A}_k^2 = -\frac{\partial H}{\partial A_{-k}^1} = -\frac{\partial W}{\partial A_{-k}^1} \ . \tag{7}$$

The kinetic energy K is diagonal,

$$K = \tfrac{1}{2}\sum_k A_{-k}^2 A_k^2 I_k \tag{8}$$

with

$$I_k = \left[\frac{2}{m}\right][1 - \cos(k)] \ . \tag{9}$$

In what follows, the average $\langle A \rangle$ of any function of the generalized coordinates and momenta is defined as

$$\langle A \rangle = \frac{\mathrm{Tr}[A\exp(-\beta H)]}{\mathrm{Tr}\exp(-\beta H)} \ ,$$

where Tr denotes integration over the phase space of the system and $\beta = 1/k_B T$, where k_B is Boltzmann's constant.

In order to calculate Kubo's response functions[16]

$$-\langle\{A_k^{\alpha}(t), A_{-q}^{\beta}(0)\}\rangle \quad (\alpha,\beta = 1,2) \ , \tag{10}$$

we derive a general representation for the Poisson brackets[17] (PB)

$$-\phi_{kq}^{\alpha\beta}(t) \equiv \{A_k^{\alpha}(t), A_{-q}^{\beta}(0)\} = \sum_p \left[\frac{\partial A_k^{\alpha}(t)}{\partial A_p^{1}} \frac{\partial A_{-q}^{\beta}(0)}{\partial A_{-p}^{2}} - \frac{\partial A_k^{\alpha}(t)}{\partial A_{-p}^{2}} \frac{\partial A_{-q}^{\beta}(0)}{\partial A_p^{1}} \right] . \tag{11}$$

If we differentiate (11) and use (6) and (7) together with basic properties of PB we obtain ($\beta = 1,2$)

$$\dot{\phi}_{kq}^{1\beta}(t) = -I_k\{A_k^2(t), A_{-q}^{\beta}(0)\} = I_k \phi_{kq}^{2\beta}(t) \ , \tag{12}$$

$$\dot{\phi}_{kq}^{2\beta}(t) = \left\{ + \frac{\partial W}{\partial A_{-k}^{1}}, A_{-q}^{\beta}(0) \right\} = - \sum_p \frac{\partial^2 W}{\partial A_{-k}^{1}(t) \partial A_p^{1}(t)} \phi_{pq}^{1\beta}(t) \ . \tag{13}$$

Introducing the matrices

$$\phi(t) = [\phi_{kq}^{\alpha\beta}(t)], \quad h(t) = [h_{kq}^{\alpha\beta}(t)] \tag{14a}$$

with

$$\begin{aligned} h_{kq}^{21} &= - \frac{\partial^2 W}{\partial A_{-k}^{1} \partial A_q^{1}} , \\ h_{kq}^{12} &= I_k \delta_{kq} \ , \\ h_{kq}^{22} &= h_{kq}^{11} = 0 \ , \end{aligned} \tag{14b}$$

Eqs. (12) and (13) may be written in compact form:

$$\dot{\phi}_{kq}^{\alpha\beta}(t) = \sum_{p,\gamma} h_{kp}^{\alpha\gamma}(t) \phi_{pq}^{\gamma\beta}(t) \ . \tag{15}$$

δ_{kq} denotes the Kronecker symbol: $\delta_{kk} = 1$, $\delta_{kq} = 0$ for $k \neq q$.

The solution of the first-order linear differential equations (15) is given by

$$\phi_{kq}^{\alpha\beta}(t) = \left\{ \left[T \exp\left[\int_0^t h(\tau) d\tau \right] \right] \phi(0) \right\}_{kq}^{\alpha\beta} \tag{16}$$

where we have introduced the usual T ordering ($T \cdots$) which determines the order of the matrix multiplication appearing in (16) (latest time to the left). Equation (16) gives the desired general representation of the Poisson brackets (11).

Before developing our theory further let us point out some general relations for the response functions (10), which mainly follow from the symmetry properties of the system.[16] $\langle\phi_{kq}^{\alpha\beta}(t)\rangle$ is real and diagonal in k,q because of translational invariance of the lattice:

$$\langle\phi_{kq}^{\alpha\beta}(t)\rangle = -\delta_{kq}\langle\{A_k^{\alpha}(t), A_{-k}^{\beta}(0)\}\rangle \ . \tag{17}$$

Equation (6) together with time translational invariance result in the following relations which connect the various components of the response functions:

$$-\langle\phi_{kk}^{12}(t)\rangle = +\langle\phi_{kk}^{21}(t)\rangle = \left[\frac{1}{I_k}\right] \frac{d}{dt}\langle\phi_{kk}^{11}(t)\rangle \ , \tag{18}$$

$$-\langle\phi_{kk}^{22}(t)\rangle = \left[\frac{1}{I_k^2}\right] \frac{d^2}{dt^2}\langle\phi_{kk}^{11}(t)\rangle \ . \tag{19}$$

The response functions $\langle\phi_{kk}^{\alpha\alpha}(t)\rangle$ are odd in time:

$$\langle\phi_{kk}^{\alpha\alpha}(t)\rangle = -\langle\phi_{kk}^{\alpha\alpha}(-t)\rangle \ .$$

Coming back now to our main line of argument we perform the thermal average from Eqs. (16). $\phi_{kq}^{\gamma\beta}(0)$ is calculated from Eq. (11):

$$\begin{aligned} &\phi_{kq}^{11}(0) = \phi_{kq}^{22}(0) = 0 \ , \\ &-\phi_{kq}^{12}(0) = \phi_{kq}^{21}(0) = \delta_{kq} \ . \end{aligned} \tag{20}$$

Thus we obtain the following explicit representations for Kubo's response functions:

$$\begin{aligned} &\langle\phi_{kk}^{11}(t)\rangle = U_k^{12}(t), \quad \langle\phi_{kk}^{12}(t)\rangle = -U_k^{11}(t) \ , \\ &\langle\phi_{kk}^{21}(t)\rangle = U_k^{22}(t), \quad \langle\phi_{kk}^{22}(t)\rangle = -U_k^{21}(t) \ , \end{aligned} \tag{21}$$

with

$$U_k^{\alpha\beta}(t) = \left\langle \left[T \exp\left[\int_0^t d\tau h(\tau) \right] \right]_{kk}^{\alpha\beta} \right\rangle . \tag{22}$$

Taking into account Eqs. (18), (19), and (21) the matrix $U_k(t) = [U_k^{\alpha\beta}(t)]$ can be written as

$$U_k(t) = [U_k^{\alpha\beta}(t)] = \begin{bmatrix} \frac{1}{I_k}\dot{U}_k^{12}(t) & U_k^{12}(t) \\ \frac{1}{I_k^2}\ddot{U}_k^{12}(t) & \frac{1}{I_k}\dot{U}_k^{12}(t) \end{bmatrix} . \tag{23}$$

All functions appearing in (21) and (23) are expressed by $\langle \phi_{kk}^{11}(t) \rangle = U_k^{12}(t)$, which is the response function of relative displacements transformed to wave-vector space. Equation (22) together with (14a) and (14b) gives a general expression for this function, which is valid for any NN interaction potential $\phi(x_i - x_{i-1})$.[18] The response of the system is determined by correlations of the second derivatives of the potential energy with respect to the coordinates R_j, i.e., correlations of the force constants for instantaneous particle positions.

III. THE EXPONENTIAL LATTICE IN LOW-TEMPERATURE APPROXIMATION

Proceeding from the general representation (22), we work out an approximation scheme valid at low temperatures. Firstly, we perform the lowest-order decoupling approximation with respect to $h(t)$. We obtain from (22) for $\alpha = 1, \beta = 2$:

$$U_k^{12}(t) = \left[\exp \left[\int_0^t \langle h(\tau) \rangle d\tau \right] \right]_{kk}^{12} . \tag{24}$$

The approximation of the response function described by this formula corresponds to the renormalized harmonic approximation. This result suggests an expansion in terms of cumulants of $h_{kp}^{\alpha\beta}(t)$.

Applying Kubo's generalized cumulant expansion[19] we obtain from (22)

$$U_k(t) = [TV(t,0)]_{kk} , \tag{25}$$

where we have defined

$$V(t,0) = \exp \left[\int_0^t \langle h(t_1) \rangle dt_1 + \int_0^t dt_1 \int_0^{t_1} dt_2 \langle h(t_1) h(t_2) \rangle_c + \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \langle h(t_1) h(t_2) h(t_3) \rangle_c + \cdots \right] . \tag{26}$$

The symbol $\langle \cdots \rangle_c$ signifies the cumulant average with respect to the matrix elements of h. This averaging operation preserves the matrix character of $h(t)$. Thus the T symbol in (25) must be retained to determine the order of the matrix multiplications in the exponential series (26).

At low temperatures cumulants higher than second order in $h_{kq}^{\alpha\gamma}$ may be neglected.[20] Thus we obtain from (26)

$$V(t,0) = \exp \left[\int_0^t \langle h(t_1) \rangle dt_1 + \int_0^t dt_1 \int_0^{t_1} dt_2 \langle h(t_1) h(t_2) \rangle_c \right] . \tag{27}$$

We shall now show this equation leads to an integrodifferential equation for the determination of $U_k^{12}(t)$ at low temperatures.

First we form the derivative of Eq. (25) with (27):

$$\dot{U}_k(t) = \sum_p \langle h(t) \rangle_{kp} [TV(t,0)]_{pk} + \int_0^t d\tau \sum_p \langle h_{kp}(t) [TV(t,0) h(\tau)]_{pk} \rangle_c . \tag{28}$$

For $0 < \tau < t$ holds

$$\int_0^t dt_1 \int_0^{t_1} dt_2 \cdots = \int_0^\tau dt_1 \int_0^{t_1} dt_2 \cdots + \int_\tau^t dt_1 \int_\tau^{t_1} dt_2 \cdots + \int_\tau^t dt_1 \int_0^\tau dt_2 \cdots .$$

One can therefore write the expression (27) in the following way:

$$V(t,0) = V(t,\tau) V(\tau,0) \exp \left[\int_\tau^t dt_1 \int_0^\tau dt_2 \langle h(t_1) h(t_2) \rangle_c \right] . \tag{29a}$$

We will see below that $\langle h(t_1) h(t_2) \rangle_c$ is proportional to $k_B T$ at low temperatures. Thus in this region the exponential in Eq. (29) may be set at unity and the time-ordered product $[TV(t,0) h(\tau)]$ in Eq. (28) may be approximated as[21]

$$[TV(t,0) h(\tau)] = [TV(t,\tau)] h(\tau) [TV(\tau,0)] , \tag{29b}$$

known as the "disentangling" approximation.[22]

Following this procedure with (28) we obtain

$$\dot{U}_k^{\alpha\beta}(t) = \sum_\gamma \langle h(t)_{kk}^{\alpha\gamma} \rangle U_k^{\gamma\beta}(t) + \int_0^t d\tau \sum_{\substack{p,\gamma \\ \delta,\nu}} \langle h_{kp}^{\alpha\gamma}(t-\tau) U_p^{\gamma\delta}(t-\tau) h_{pk}^{\delta\nu}(0) U_k^{\nu\beta}(\tau) \rangle_c . \tag{30}$$

In this reduction we used the diagonality of $[TV(t,0)]$ in wave-vector space, $[TV(\tau,0)]_{qk}^{\nu\beta} = \delta_{qk} \times U_k^{\nu\beta}(\tau)$, together with time translational invariance.

The quantities $U_p^{\gamma\delta}$ in (30) are constants in phase space. The only nonvanishing cumulant $\langle h_{kp}^{\alpha\gamma}(t-\tau) h_{pk}^{\delta\nu}(0)\rangle_c$ is the one with $\alpha = \delta = 2$, $\gamma = \nu = 1$. This is a direct consequence of the definition (14b), only the matrix elements h_{kq}^{21} may be nontrivial functions in phase space. Therefore Eqs. (30) are reduced to the following set of equations ($\beta = 1,2$):

$$\dot{U}_k^{1\beta}(t) = I_k U_k^{2\beta}(t) \ , \tag{31}$$

$$\dot{U}_k^{2\beta}(t) = \langle h_{kk}^{21}\rangle U_k^{1\beta}(t) - \int_0^t d\tau\, m_k(t-\tau) U_k^{1\beta}(\tau) \ , \tag{32}$$

where the kernel $m_k(s)$ is defined as

$$m_k(s) = -\sum_p \langle h_{kp}^{21}(s) h_{pk}^{21}(0)\rangle_c U_p^{12}(s) \ . \tag{33}$$

A further reduction of this set of Eqs. (31) and (32) together with (33) may be made. First we note that the two equations (31) ($\beta = 1,2$) are already contained in (23). Then, utilizing (31) we eliminate $U^{2\beta}$ from (32) and obtain

$$\frac{\ddot{U}_k^{1\beta}(t)}{I_k} = \langle h_{kk}^{21}\rangle U_k^{1\beta}(t) - \int_0^t dt_1 m_k(t-t_1) U_k^{1\beta}(t_1) \ . \tag{34}$$

We see that $U_k^{11}(t)$ and $U_k^{12}(t)$ are solutions of the same differential equation, differing only with respect to the initial conditions. From (22) and (23) these initial conditions are given by

$$U_k^{11}(0) = 1, \quad \dot{U}_k^{11}(0) = 0 \ , \tag{35a}$$

$$U_k^{12}(0) = 0, \quad \dot{U}_k^{12}(0) = I_k \ . \tag{35b}$$

Applying (34) for $\beta = 2$ we see that the two functions $U_k^{11}(t)$ and $\dot{U}_k^{12}(t)/I_k$ coincide with respect to the initial conditions:

$$U_k^{11}(0) = \frac{\dot{U}_k^{12}(0)}{I_k} \ ,$$

$$\left[\frac{d}{dt} U_k^{11}(t)\right]_{t=0} = \left[\frac{d}{dt}\frac{\dot{U}_k^{12}(t)}{I_k}\right]_{t=0} \ .$$

The two functions are, in fact, identical. This follows from (34) for $\beta = 2$ by differentiation: $\dot{U}_k^{12}(t)/I_k$ and $U_k^{11}(t)$ are solutions of the same differential equation. Thus we obtain

$$U_k^{11}(t) = \frac{\dot{U}_k^{12}(t)}{I_k} \ . \tag{36}$$

It should be noted at this point that all the exact equations summarized in (23) are satisfied by our approximate solution for low temperatures. This solution is obtained by the second-order cumulant approach together with the disentangling approximation (29b). Thus the approximation procedure is consistent with the symmetry properties of the system.

The response function $U_k^{12}(t)$ is the solution of the integrodifferential equations (34) and (33) with initial condition (35b), and can be determined if the cumulants $\langle h_{kk}^{21}\rangle$ and $\langle h_{kp}^{21}(s) h_{pk}^{21}(0)\rangle_c$ in Eq. (33) are known. In the special case of an exponential interaction potential (2), these cumulants can easily be calculated. Following Eqs. (14b), (4), and (5), h_{kp}^{21} is given by

$$h_{kp}^{21} = -\frac{\partial^2 W}{\partial A_{-k}^{1}\partial A_p^{1}} = -\frac{1}{M}\sum_{n=1}^{M} \exp[i(p-k)n]\phi''(R_n) . \tag{37}$$

According to Eqs. (2) and (1) for the Toda lattice,

$$\phi''(R_n) = ab + b\dot{S}_n \tag{38}$$

holds, so that we obtain

$$h_{kp}^{21}(t) = -ab\delta_{kp} - (b/\sqrt{M})\dot{A}_{k-p}^{2}(t) \ . \tag{39}$$

With this expression for $h_{kp}^{21}(t)$ we can reduce the cumulants $\langle h_{kk}^{21}\rangle$ and $\langle h_{kp}^{21}(t) h_{pk}^{21}(0)\rangle_c$ to known quantities:

$$\langle h_{kk}^{21}\rangle = -ab \ , \tag{40}$$

$$\langle h_{kp}^{21}(t) h_{pk}^{21}(0)\rangle_c = -\frac{b^2}{M}\frac{d^2}{dt^2}\langle A_{k-p}^{2}(t) A_{p-k}^{2}(0)\rangle . \tag{41}$$

The frequencies

$$\omega_k = (-I_k\langle h_{kk}^{21}\rangle)^{1/2} = (abI_k)^{1/2}$$

in Eq. (34) are identical with those obtained from the self-consistent phonon theory. For the Toda lattice the frequencies are temperature independent at a given pressure ($p = 0$ in our case). According to Eq. (41) the second-order cumulants are proportional to the correlations of the generalized forces $\dot{A}_{k-p}^{2}$:

$$\langle \dot{A}_{k-p}^{2}(t)\dot{A}_{p-k}^{2}(0)\rangle = -\frac{d^2}{dt^2}\langle A_{k-p}^{2}(t) A_{p-k}^{2}(0)\rangle \ . \tag{42}$$

Employing Eq. (6) and the fluctuation-dissipation theorem[16] in its classical version,

$$\beta\langle \dot{A}^1_{k-p}(t)A^1_{p-k}(0)\rangle = -\,U^{12}_{k-p}(t)\ ,$$

we obtain from Eq. (41)

$$\langle h^{21}_{kp}(t)h^{21}_{pk}(0)\rangle_c = -\,\frac{k_BTb^2}{MI^2_{k-p}}\ddot{U}^{12}_{k-p}(t)\ . \tag{43}$$

Equation (43) does not make sense for $k-p=0$. The right-hand side is not defined for $k=p$ [$\ddot{U}^{12}_0(t)/I^2_0 = 0/0$]. The corresponding contribution to the kernel (33) is of order $1/M$. We shall neglect it, assuming that it is sufficiently small in our calculations. This restricts the applicability of the approximation procedure developed to systems with a very large number of particles.[23]

The final equations for the determination of the response functions $U^{12}_k(t)$ can be formulated as follows:

$$\frac{\ddot{U}^{12}_k(t)}{I_k} + abU^{12}_k(t) + \int_0^t d\tau\, m_k(t-\tau)U^{12}_k(\tau) = 0 \tag{44}$$

with

$$m_k(s) = \frac{k_BTb^2}{M}\sum_{p\neq k}\frac{1}{I^2_{k-p}}U^{12}_p(s)\ddot{U}^{12}_{k-p}(s)\ . \tag{45}$$

They are valid for the Toda lattice in the low-temperature region.

A few summarizing remarks may be added. The response of the generalized coordinate A^1_k at time t (>0) in reaction to a small perturbation $A^{\beta}_{-q}(0)\delta(t)$ which contributes only at time $t=0$ is given by the Poisson bracket

$$\delta A^1_k(t) = \{A^1_k(t), A^{\beta}_{-q}(0)\} = -\,\phi^{1\beta}_{kq}(t)\ .$$

The quantity $\delta A^1_k(t)$ represents the change in the variable $A^1_k(t)$ with respect to the value in which it evolves due to the natural motion of the system. Once this deviation has been set up, the system reacts with the generation of a restoring force $-\sum_p h^{21}_{kp}\phi^{1\beta}_{pq}$ which is calculated according to classical mechanics. Following Newton's law this force determines the time evolution of the quantity $\delta A^1_k(t)$. The corresponding dynamical equations are summarized in (15). The solution (16) of these equations allows one to calculate the thermal average $\langle \delta A^1_k(t)\rangle = -\langle \phi^{1\beta}_{kq}(t)\rangle$ in terms of correlations of the instantaneous force constants of the system. In the present case of the exponential lattice, the force constants can be expressed by the forces themselves, i.e., the second time derivatives of the generalized coordinates. Therefore, the correlations of the instantaneous force constants are given by the fourth time derivatives of the correlations of the generalized coordinates. In this way we obtain the self-consistent system of Eqs. (44) and (45) to determine $U^{12}_k(t)$.

IV. CONCLUSIONS

This paper contributes to the theory of thermally excited phonon systems with strong nonlinear characteristics. The response functions corresponding to the canonically conjugate variables of the system have been represented in terms of correlations of the force constants of the lattice (defined with respect to instantaneous particle positions). A calculation of the response functions generally implies an approximate treatment of these correlations, e.g., a decoupling procedure. In the special case of an exponential restoring force between adjacent particles the force constants corresponding to instantaneous particle positions can be expressed by time derivatives of the canonical momenta themselves. This result again illuminates the role of the linear exponential lattice within the theory of strong anharmonic phonon systems: The time-dependent response functions of the lattice are determined by correlations of the forces themselves. A self-consistent system of equations is valid at low temperatures which allows a calculation of the response functions of the thermalized system.

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[20]nth-order cumulants may be estimated by their equal-time values. Because displacements $r_j = R_j - D$ at different sites of the lattice are uncorrelated at equal time, third-order cumulants (to give an example) contribute according to the formula

$$\langle h(0)h(0)h(0)\rangle_c \sim \langle \phi''(R)\phi''(R)\phi''(R)\rangle_c$$
$$\approx (ab)^3[\langle \exp(-3br)\rangle - 3\langle \exp(-2br)\rangle + 2] .$$

This is a direct implication from Eqs. (2) and (37). We calculate the thermal averages $\langle \exp(-nbr)\rangle$ with the help of the partition function (Ref. 7) and obtain

$$\langle h(0)h(0)h(0)\rangle_c \sim 2(ab)^3(k_B T b/a)^2 ,$$
$$\langle h(0)h(0)h(0)\rangle_c \sim (ab)^2 k_B T b/a .$$

Thus higher-order cumulants lead to higher-order contributions with respect to the temperature.
[21]Corrections to this approximation can be obtained by a Taylor expansion of the exponential in Eq. (29a). The corresponding additional terms to (29b) provide higher-order contributions with respect to temperature.
[22]M. Blume and J. Hubbard, Phys. Rev. B 1, 3815 (1970)
[23]We have formulated the periodicity condition in the following way: $S_l = S_{M+l}$, $R_l = R_{m+l}$ for any index l. The periodicity condition at defined periodicity length L is given by $X_{M+l} - X_l = L$. It corresponds to a ring of M particles with the total length L. The displacements $R_1, \ldots, R_M$ in this case are not dynamically independent ($\sum_{n=1}^{M} R_n = L$). The calculations will be only slightly different, resulting in a different value of the term $p = k$ which contributes to the kernel $m_k(s)$. Again, however, the final equations (44) and (45) remain valid if there are sufficient particles in the ring so that we can neglect the contribution $p = k$.