

Quantum theory of nonlinear oscillators interacting with the medium

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The time correlation function $Q_K(t)$ of selected (e.g. local or quasilocal) nonlinear oscillations interacting with the medium and the spectral representation $Q_K(\omega)$ are determined. The general case of an arbitrary relation between nonequidistance of the vibrational state levels, due to nonlinearity, and their inverse lifetime, due to interaction with the medium, is considered. The problem is solved by means of an integral operator equation which reduces to a partial differential equation. It is shown that asymptotically for large times, $Q_K(t)$ can be expressed in elementary functions and $Q_K(\omega)$ in quadratures. The shape of the spectral distribution and its fine structure are investigated in various cases.

1. INTRODUCTION

The quantum theory of a harmonic oscillator interacting with the medium was developed in a number of papers^[1-4]. The kinetics of the variation of the quantum states of the oscillator was investigated in detail and the time-dependent correlation functions of its coordinates were determined. The results obtained for harmonic (linear) oscillators can, however, be modified in many respects on going over to nonlinear detached oscillators. The reason is that the levels of nonlinear oscillators become non-equidistant. Therefore, neglecting the interaction with the medium, the spectrum of the absorption or scattering of waves by a nonlinear oscillator will consist of a set of lines corresponding to quantum transitions from different levels and separated by an amount $\sim \Delta\omega$ that characterizes the degree of nonlinearity (and not of one line, as in the case of a linear oscillator).

Allowance for the interaction with the medium, just as for the linear oscillator, leads to quantum transitions between levels, to a finite lifetime τ of the states, and to line broadening. Depending on the relation between τ^{-1} and $\Delta\omega$, the indicated set of lines will make up a somewhat smeared fine structure or else will coalesce into a single broadened distribution. The shape of the resultant spectral distribution was considered in a number of papers^[5,6] by the Green's function method, but only the limiting cases $\Delta\omega \ll \tau^{-1}$ and $\tau^{-1} = 0$ could be investigated. The use of the asymptotic methods of nonlinear mechanics and of certain results of the theory of random processes has also made it possible to construct a complete classical theory of the spectral distribution of nonlinear oscillations^[7].

The classical theory, however, is valid only at high temperatures and does not make it possible to investigate the fine structure—which is purely quantum in nature—of the spectral distribution. A quantum theory is needed to investigate the fine structure at low temperatures. Such a quantum theory, valid for arbitrary relations between $\Delta\omega$ and τ^{-1} (but under the assumption that $\Delta\omega$ and τ^{-1} are small in comparison with the frequencies ω_K of the detached oscillators), will be developed below with the aid of a special method based on the use of an operator integral equation that reduces to a partial differential equation that can be solved in terms of elementary functions. In some respects, it is simpler than the classical theory.

2. FORMULATION OF PROBLEM

We consider the time-dependent correlation function $Q_K(t)$ of the annihilation and creation operators a_K and a_K^+ of detached oscillators κ , and its spectral representation $Q_K(\omega)$:

$$Q_\kappa(t) = \langle a_\kappa(t) a_\kappa^+(0) \rangle = Z^{-1} \text{Sp} (a_\kappa^+ e^{-\lambda H} e^{iHt} a_\kappa e^{-iHt}), \quad (1)$$

$$Q_\kappa(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} Q_\kappa(t) e^{i\omega t} dt = \frac{1}{\pi} \int_0^{\infty} \text{Re} Q_\kappa(t) e^{i\omega t} dt.$$

Here $\hbar = 1$, $Z = \text{Tr} \exp(-\lambda H)$, $\lambda = (kT)^{-1}$, $a_K = a_K(0)$, and H is the Hamiltonian of the system.

If, for example, the considered detached oscillations are local or quasilocal vibrations in crystals, then $Q_K(\omega)$ determines, apart from factors that depend little on the frequency, the spectral distribution of the absorption or scattering of infrared radiation or inelastic scattering of neutrons. It is precisely the function $Q_K(\omega)$ that can change qualitatively when nonlinearity effects are taken into account. The nonlinearity gives rise to only small corrections, $\sim \Delta\omega/\omega_K$, to the correlation function $\langle \hat{n}_K(t) \hat{n}_K(0) \rangle$ ($\hat{n}_K = a_K^+ a_K$), and also to the probabilities of the transitions between levels.

We choose as the model of the medium a system of harmonic oscillations k with frequencies ω_k belonging to a continuous spectrum. The Hamiltonian of the detached oscillations interacting with such a medium is expressed in the form

$$H = H_0 + H_i, \quad H_i = \sum_k H_k(a_k + a_k^+) + \frac{1}{2} \sum_{kk'} H_{kk'}(a_k + a_k^+)(a_{k'} + a_{k'}^+),$$

$$H_0 = \sum_x \omega_x \hat{n}_x + \frac{1}{2} \sum_{xx'} V_{xx'} \hat{n}_x \hat{n}_{x'} + \sum_k \omega_k \hat{n}_k,$$

$$H_k = \sum_x V_{xk}(a_x + a_x^+) + \sum_{xk'} V_{xk'k}(a_x + a_x^+)(a_{k'} + a_{k'}^+), \quad (2)$$

$$H_{kk'} = \sum_x V_{xkk'}(a_x + a_x^+) + \sum_{xx'} V_{xx'kk'}(a_x + a_x^+)(a_{k'} + a_{k'}^+),$$

$$\hat{n}_k = a_k^+ a_k, \quad \hat{n}_x = a_x^+ a_x.$$

This Hamiltonian is a satisfactory model for the description of local and quasilocal vibrations of weakly bound impurity atoms in crystals. It can, however, be used to investigate the considered effects also in a more general case of such vibrations in defects with inversion centers (and also in other systems with detached oscillations). We note that we have left out of

the Hamiltonian (2) certain "nonresonant" terms (for example, of the type $a_k^3 a_k^\dagger$), which lead to corrections of higher order of smallness.

We confine ourselves henceforth to the case of centers of low symmetry, in which the frequencies of the detached oscillations do not coincide ($\omega_K \neq \omega_{K'}$) and are not close to each other. In addition, we exclude the cases of resonant situations of the type $\omega_K \approx \omega_{K_1} \pm \omega_{K_2}$ or $\omega_K \pm \omega_{K'} \approx \omega_{K_1} \pm \omega_{K_2}$. The nonlinearity of the oscillator is assumed to be small ($|V_{KK'}| \ll \omega_K$).

It is convenient to calculate the trace in formula (1) with the aid of the complete system of the eigenfunctions $(n_K|$ and $(n_K|$ (or $(m_K|$ and $(m_K|$) of the operators \hat{n}_K and \hat{n}_K for the detached oscillations and for the continuous-spectrum oscillations. We change over to the interaction representation, i.e., we introduce the operator

$$U(t) = e^{iH_0 t} e^{-iHt} = T \exp \left\{ -i \int_0^t H_i(\tau) d\tau \right\}, \quad H_i(\tau) = e^{iH_0 \tau} H_i e^{-iH_0 \tau}, \quad (3)$$

where T is the chronological-ordering symbol. Then expression (1) for $Q_K(t)$ takes the form

$$\begin{aligned} Q_K(t) &= \exp[-i(\omega_K - 1/2 V_{KK})t] \exp(\lambda \omega_K) \bar{Q}_K(t), \\ \bar{Q}_K(t) &= \frac{1}{Z_i} \sum_{\dots, m_{K'}, \dots} m_{K'} F(m_{K'}, t) \exp\left(-\lambda \sum_{K'} m_{K'} \omega_{K'} - i \sum_{K'} V_{KK'} m_{K'} t\right), \\ F(m_{K'}, t) &= \sum_{\dots, n_{K'}, \dots} \sqrt{n_{K'}/m_{K'}} F(n_{K'}, m_{K'}), \\ Z_i &= \sum_{\dots, n_{K'}, \dots} \exp\left(-\lambda \sum_{K'} n_{K'} \omega_{K'}\right), \\ F(n_{K'}, m_{K'}) &= \frac{1}{Z_i} \sum_{\dots, n_K, m_K, \dots} (n_{K'} - \delta_{KK'}, n_K | U^{-1}(t + i\lambda) | m_{K'} - \delta_{KK'}, m_K) \cdot \\ &\quad \times (m_{K'}, m_K | U(t) | n_{K'}, n_K) \exp\left(-\lambda \sum_{K'} m_{K'} \omega_{K'}\right), \\ Z_i &= \sum_{\dots, m_{K'}, \dots} \exp\left(-\lambda \sum_{K'} m_{K'} \omega_{K'}\right). \end{aligned} \quad (4)$$

We have discarded here small terms of the order of

$$\lambda |V_{KK'}| \bar{n}_{K'} < |V_{KK'}| \omega_{K'}^{-1} \ll 1 \quad (\bar{n}_K = [e^{\lambda \omega_K} - 1]^{-1}).$$

The operator $U^{-1}(t + i\lambda) = U^\dagger(t - i\lambda)$ can be calculated by a formula similar to (3), in which T is replaced by the operator T^- , which carries out the chronological ordering in the opposite direction, and in which the sign of the argument of the exponential is reversed and t is replaced by $t + i\lambda$.

To calculate $\bar{Q}_K(t)$, we exclude the oscillations of the continuous spectrum. We consider first a simple case when processes in which two oscillations of the continuous spectrum participate can be neglected in the Hamiltonian (2), $H_{KK'} = 0$. The analysis is then generalized to include the case $H_{KK'} \neq 0$.

In this case $U(t)$ (or $U^{-1}(t + i\lambda)$) breaks up into a τ -ordered product (with respect to k) of factors of the type

$$\exp \left\{ -i \int_0^t H_k(\tau) [a_k(\tau) + a_k^\dagger(\tau)] d\tau \right\}. \quad (5)$$

The operators $H_k(\tau)$ contain the small factor $(1/N)^{1/2}$, where N is the number of atoms of the system. This enables us to confine ourselves to quadratic terms in the expansion of the exponential factor (5), to calculate directly the matrix elements on the functions $(n_K|$ and

$(m_K|$, and to carry out statistical averaging over the occupation numbers m_K . Then, reducing the product over k to an exponential function of a sum over k , we obtain the following expression for $F(n_{K'}, m_{K'})$:

$$\begin{aligned} F(n_{K'}, m_{K'}) &= (n_{K'} - \delta_{KK'}, m_{K'} | T T^- \exp G | m_{K'} - \delta_{KK'}, n_{K'}), \\ G &= \sum_k \left\{ \int_0^{t+i\lambda} \int_0^t \bar{H}_k(\tau_1) H_k(\tau_2) \varphi_k(\tau_2 - \tau_1) d\tau_1 d\tau_2 - \right. \\ &\quad \left. - \int_0^{t+i\lambda} \int_0^{\tau_1} \bar{H}_k(\tau_2) \bar{H}_k(\tau_1) \varphi_k(\tau_2 - \tau_1) d\tau_1 d\tau_2 - \int_0^{\tau_1} \int_0^{\tau_1} H_k(\tau_1) H_k(\tau_2) \varphi_k(\tau_1 - \tau_2) d\tau_1 d\tau_2 \right\}, \\ \varphi_k(\tau) &= \bar{n}_k e^{i\omega_k \tau} + (\bar{n}_k + 1) e^{-i\omega_k \tau}, \quad \bar{n}_k = (e^{\lambda \omega_k} - 1)^{-1}. \end{aligned} \quad (6)$$

We have introduced here a symbolic notation according to which the operators \bar{H}_K (with superior bar) are expressed as matrices of the first functions in the matrix brackets to the left and to the right of the operators, in our case the functions $(n_{K'} - \delta_{KK'}|$ and $|m_{K'} - \delta_{KK'}|$, and the operators H_K (without the superior bar) are expressed in terms of the second functions, in our case on the functions $(m_{K'}|$ and $|n_{K'}|$. The ordering operators T and T^- pertain respectively to $H_K(\tau)$ and $\bar{H}_K(\tau)$.

In the derivation of (6), we used only the transition to the limit $N \rightarrow \infty$, and these formulas are valid independently of the smallness of the interaction of the detached oscillations with the oscillations of the continuous spectrum. In the more general case, when $H_{KK'} \neq 0$, analogous expressions can be obtained by assuming that this interaction is small and the $H_{KK'}$ contain a small parameter. Calculation shows that $F(n_{K'}, m_{K'})$ is also determined in this case by formula (6), and in second order in the interaction constants (in both H_K and $H_{KK'}$) it is necessary to add to the term G in (6) a term G' equal to

$$\begin{aligned} G' &= -\frac{1}{2} i \sum_k (2\bar{n}_k + 1) \left[\int_0^t H_{kk}(\tau) d\tau - \int_0^{t+i\lambda} \bar{H}_{kk}(\tau) d\tau \right] \\ &\quad + \frac{1}{2} \sum_{K, K'} \left\{ \int_0^{t+i\lambda} \int_0^t \bar{H}_{Kk'}(\tau_1) H_{Kk'}(\tau_2) \varphi_{Kk'}(\tau_2 - \tau_1) d\tau_1 d\tau_2 \right. \\ &\quad \left. - \int_0^{t+i\lambda} \int_0^{\tau_1} \bar{H}_{Kk'}(\tau_2) \bar{H}_{Kk'}(\tau_1) \varphi_{Kk'}(\tau_2 - \tau_1) d\tau_1 d\tau_2 \right. \\ &\quad \left. - \int_0^{\tau_1} \int_0^{\tau_1} H_{Kk'}(\tau_1) H_{Kk'}(\tau_2) \varphi_{Kk'}(\tau_1 - \tau_2) d\tau_1 d\tau_2 \right\}; \end{aligned} \quad (7)$$

$$\begin{aligned} \varphi_{Kk'}(\tau) &= \bar{n}_k \bar{n}_{k'} \exp\{i(\omega_k + \omega_{k'})\tau\} + (1 + \bar{n}_k)(1 + \bar{n}_{k'}) \exp\{-i(\omega_k + \omega_{k'})\tau\} \\ &\quad + 2(\bar{n}_k + 1)\bar{n}_{k'} \exp\{-i(\omega_k - \omega_{k'})\tau\}. \end{aligned}$$

3. DETERMINATION OF THE TIME-DEPENDENT CORRELATION FUNCTIONS OF THE DETACHED SELECTED OSCILLATIONS

We determine the correlation function $\bar{Q}_K(t)$ from formulas (4) and (6) first for the simplest case, when $H_{KK'} = 0$ in the interaction Hamiltonian, and the coefficients $V_{KK'k}$ vanish in H_K , i.e.,

$$H_k = \sum_x V_{xk} (a_x + a_x^\dagger).$$

This case corresponds, for example, to nonlinear quasilocal vibrations of a weakly bound impurity atom or molecule in the case of harmonic interaction with the oscillations of a continuous spectrum.

The region of the peak of the spectral distribution $Q_K(\omega)$ is determined by the behavior of the function $Q_K(t)$ (or $\bar{Q}_K(t)$) at long times, considerably longer than the period of the selected oscillations and of the continuous-spectrum oscillations:

$$t \gg t_0, \quad t_0 = \max(\omega_m^{-1}, \omega_k^{-1})$$

(ω_m is the maximum oscillation frequency in the continuous spectrum). The calculation will therefore be performed from now on for the asymptotic case of such long times. Accordingly, we shall take into account in the formulas the terms of the type $\epsilon t/t_0$, where ϵ are small constants due to nonlinearity and interaction with the medium, and small terms $\sim \epsilon$ are not taken into account. This means that we investigate the shape of the spectral distribution of the peak $Q_K(\omega)$, but not the attenuation of its intensity (which is small in the considered case of small ϵ).

The functions $F(m_{K'}, t)$ determined by formulas (4) and (6) can be calculated by expanding $\exp G$ in (6). The first two terms of the expansion, according to (6) and (4), are given by

$$\begin{aligned} F(m_{K'}, t) &= 1 + F^{(1)}(m_{K'}, t), \quad F^{(1)}(m_{K'}, t) = -b_K(m_{K'})t \\ &+ 2 \sum_{x_i} \Gamma_{x_i}(m_{x_i} - \delta_{x_{K'}}) (\bar{n}_{x_i} + 1) \int_0^t \exp(iV_{x_{K'}}\tau) d\tau \\ &+ 2 \sum_{x_i} \Gamma_{x_i}(m_{x_i} + 1) \bar{n}_{x_i} \int_0^t \exp(-iV_{x_{K'}}\tau) d\tau \quad (t \gg t_0), \\ b_K(m_{K'}) &= 2 \sum_{x_i} \Gamma_{x_i} m_{x_i} (2\bar{n}_{x_i} + 1) + 2 \sum_{x_i \neq x} \Gamma_{x_i} \bar{n}_{x_i} - \Gamma_x + iP_x. \end{aligned} \quad (8)$$

Here Γ_K and P_K are the damping and the shift of the selected oscillation κ , due to its decay accompanied by phonon production (and calculated without allowance for the anharmonicity). For the considered interaction Hamiltonian we have

$$\Gamma_x = \pi \sum_k V_{xk}^2 \delta(\omega_x - \omega_k), \quad P_x = 2P \sum_k \frac{V_{xk}^2 \omega_k}{\omega_x^2 - \omega_k^2}. \quad (9)$$

The symbol P denotes that after changing over from the sum over k to the integral, the latter must be taken in the sense of the principal value.

In (8), taking into account the smallness of the interaction Hamiltonian H_i and the smallness of $|V_{KK'}|$, we could discard terms of order

$$\Gamma_x \lambda, \quad |V_{xK'}| \lambda, \quad \Gamma_x / \omega_{x_i}, \quad P_x / \omega_{x_i}$$

neglect the derivatives $d\Gamma_K/d\omega_K$ and $dP_K/d\omega_K$ together with $\Gamma_K t$ and $P_K t$ (it is implied that the frequencies ω_K do not lie near the singular points in the continuous spectrum), and replace $t + i\lambda$ in the limits of the integrals by t . Following these substitutions, the operator G can be represented in the form

$$G = G(t) = \int_0^t \int_0^{\tau_1} A(\tau_1, \tau_2) d\tau_1 d\tau_2,$$

$$A(\tau_1, \tau_2) = \sum_k [\bar{H}_k(\tau_1) \varphi_k(\tau_2 - \tau_1) - H_k(\tau_1) \varphi_k(\tau_1 - \tau_2)] [H_k(\tau_2) - \bar{H}_k(\tau_2)]. \quad (10)$$

In the second term of the expansion of $\exp G$

$$\begin{aligned} \frac{1}{2} G^2 &= \int_0^t \int_0^{\tau_1} A(\tau_1, \tau_2) d\tau_1 d\tau_2 \left[\int_0^{\tau_2} \int_0^{\tau_3} A(\tau_3, \tau_4) d\tau_3 d\tau_4 \right. \\ &\left. + \int_0^{\tau_1} \int_0^{\tau_2} A(\tau_3, \tau_4) d\tau_3 d\tau_4 + \int_{\tau_1}^{\tau_2} \int_{\tau_1}^{\tau_2} A(\tau_3, \tau_4) d\tau_3 d\tau_4 \right] \end{aligned} \quad (11)$$

we need retain in the considered asymptotic theory only the first term. Indeed, in the significant time region where $F^{(1)}(m_{K'}, t) \sim 1$, this term also becomes of the order of unity after TT-ordering and matrix formation. On the other hand, since $A(\tau_1, \tau_2)$ contains rapidly oscillating factors of the type $\exp i(\omega_K - \omega_{K'}) (\tau_1 - \tau_2)$, and in the second term the regions of integration with respect to τ_3 and τ_4 do not intersect, this term is of the order of $d\Gamma_K/d\omega_K \ll 1$ at these values of the time. The third term, on the other hand, turns out to be small, because the aforementioned factors oscillate rapidly at large $|\tau_1 - \tau_2|$ (no such situation arises in the first term, since the internal integral depends only on τ_2 and we can choose τ_1 to be close to τ_2).

Retaining only the first term in (11) and taking (10) into account, we obtain the integral equation

$$\frac{1}{2} G^2(t) = \int_0^t \int_0^{\tau_1} A(\tau_1, \tau_2) G(\tau_2) d\tau_1 d\tau_2.$$

We obtain in similar fashion a recurrence formula for the p -th term of the expansion of $\exp G$ and an operator integral relation for $\exp G$:

$$\begin{aligned} \frac{1}{p!} G^p(t) &= \int_0^t \int_0^{\tau_1} A(\tau_1, \tau_2) \frac{1}{(p-1)!} G^{p-1}(\tau_2) d\tau_1 d\tau_2, \\ \exp G(t) &= 1 + \int_0^t \int_0^{\tau_1} A(\tau_1, \tau_2) \exp G(\tau_2) d\tau_1 d\tau_2. \end{aligned} \quad (12)$$

From (12) and from the operator-ordering rules presented above it follows that the operators without bars in $A(\tau_1, \tau_2)$ are on the left of $G^{p-1}(\tau_2)$ or $\exp G(\tau_2)$, and the operators with the bars are on the right. It can be assumed here that the operators without the superior bar act to the left on the functions $|m_{K'}\rangle$, and the operators with the bar act to the right on the functions $|m_{K'} - \delta_{KK'}\rangle$; the functions $|n_{K'}\rangle$ and $|n_{K'} - \delta_{KK'}\rangle$, on the other hand, are not affected. As a result, it becomes easy to calculate the sums over $n_{K'}$ and to obtain an integral difference equation for the function $f(m_{K'}, t)$ defined by formulas (4) and (6):

$$\begin{aligned} F(m_{K'}, t) &= 1 - b_K(m_{K'}) \int_0^t F(m_{K'}, \tau) d\tau \\ &+ 2 \sum_{x_i} \Gamma_{x_i}(m_{x_i} - \delta_{x_{K'}}) (\bar{n}_{x_i} + 1) \int_0^t \exp(iV_{x_{K'}}\tau) F(\dots, m_{x_i} - 1, \dots, \tau) d\tau \\ &+ 2 \sum_{x_i} \Gamma_{x_i}(m_{x_i} + 1) \bar{n}_{x_i} \int_0^t \exp(-iV_{x_{K'}}\tau) F(\dots, m_{x_i} + 1, \dots, \tau) d\tau. \end{aligned} \quad (13)$$

Here $f(\dots, m_{K_1} - 1, \dots, \tau) \equiv F(m_1, m_2, \dots, m_{K_1} - 1, \dots, \tau)$.

As seen from (4), to find the sought correlation function it is necessary to know not the quantities $F(m_{K'}, \tau)$ themselves, but their sum over $m_{K'}$ with a certain weight. We therefore consider a function of continuously varying parameters $x_{K'}$

$$F(x_{K'}, t) = \sum_{\dots, m_{K'}, \dots} m_{K'} F(m_{K'}, t) \exp\left(-\sum_{K'} m_{K'} x_{K'}\right). \quad (14)$$

It follows from (13) and (14) that this function satisfies a linear partial differential equation of first order

$$\frac{\partial F(x_{K'}, t)}{\partial t} + \sum_{x_i} m_{x_i} \frac{\partial F(x_{K'}, t)}{\partial x_{x_i}} = c_{x_i} F(x_{K'}, t) \quad (15)$$

with initial condition

$$F(x_{K'}, 0) = \sum_{\dots, m_{K'}, \dots} m_{K'} \exp\left(-\sum_{K'} m_{K'} x_{K'}\right)$$

$$= (\exp(x_{\kappa}) - 1)^{-1} \prod_{\kappa'} (1 - \exp(-x_{\kappa'}))^{-1}, \quad (16)$$

We have introduced here the notation

$$\mu_{\kappa} = -2\Gamma_{\kappa} [2\bar{n}_{\kappa} + 1 - (\bar{n}_{\kappa} + 1) \exp(iV_{\kappa\kappa}t - x_{\kappa}) - \bar{n}_{\kappa} \exp(-iV_{\kappa\kappa}t + x_{\kappa})], \quad (17)$$

$$c_{\kappa} = -b_{\kappa}(0) + 2 \sum_{\kappa'} \Gamma_{\kappa'} (\bar{n}_{\kappa'} + 1) \exp(iV_{\kappa\kappa'}t - x_{\kappa'}) - 2\Gamma_{\kappa} \bar{n}_{\kappa} \exp(-iV_{\kappa\kappa}t + x_{\kappa}).$$

Equation (15) can be solved by the method of characteristics (see the Appendix). The correlation function $\bar{Q}_K(t)$ is determined, in accord with (4) and (14), by the value of $F(x_{K'}, t)$ at

$$x_{\kappa'} = \omega_{\kappa} \lambda + iV_{\kappa\kappa'} t.$$

Substituting this value, we obtain the following expression for $\bar{Q}_K(t)$:

$$\bar{Q}_{\kappa}(t) = \bar{n}_{\kappa} \Psi_{\kappa\kappa}^{-1}(t) \prod_{\kappa'} \Psi_{\kappa\kappa'}^{-1}(t) \exp[\Gamma_{\kappa} t - iP_{\kappa} \delta_{\kappa\kappa'} t + 1/2 i V_{\kappa\kappa'} (1 - \delta_{\kappa\kappa'}) t], \quad t \gg t_0. \quad (18)$$

Here

$$\Psi_{\kappa\kappa'}(t) = \text{ch } a_{\kappa\kappa'} t + \left[1 + i \frac{V_{\kappa\kappa'}}{2\Gamma_{\kappa'}} (2\bar{n}_{\kappa'} + 1) \right] \frac{\Gamma_{\kappa'}}{a_{\kappa\kappa'}} \text{sh } a_{\kappa\kappa'} t, \quad (19)$$

$$a_{\kappa\kappa'}^2 = \Gamma_{\kappa'}^2 + i\Gamma_{\kappa'} V_{\kappa\kappa'} (2\bar{n}_{\kappa'} + 1) - 1/4 V_{\kappa\kappa'}^2.$$

Formulas (4), (18), and (19) determine in explicit form the time-dependent correlation function for the considered simplest case of the interaction Hamiltonian

$$H_i = \sum_{\kappa\kappa'} V_{\kappa\kappa'} (a_{\kappa} + a_{\kappa'}^+) (a_{\kappa} + a_{\kappa'}).$$

It corresponds, for example, to nonlinear quasilocal oscillations that interact with the oscillations of the continuous spectrum (with phonons) in accordance with a harmonic law. By a perfectly similar analysis it is easy to verify that the presented formulas remain in force also in the more general case, when anharmonic interaction with phonons also plays an important role in H_i (just as in the case of local oscillations):

$$H_i = \frac{1}{2} \sum_{\kappa\kappa\kappa'} V_{\kappa\kappa\kappa'} (a_{\kappa} + a_{\kappa'}^+) (a_{\kappa} + a_{\kappa'}^+) (a_{\kappa'} + a_{\kappa'}^+),$$

The interaction is linear in the operators of the detached oscillations, but one can neglect, as before, the interaction quadratic in these operators, i.e., one can put $V_{K\kappa\kappa'} = 0$ and $V_{K\kappa\kappa'} = 0$ (see below). Allowance for the indicated terms produces in H_i only the new terms $\Gamma_{\kappa'}$ and $P_{\kappa'}$, which are added to the expressions (9) for the damping and shift of the detached oscillations:

$$\Gamma_{\kappa'} = \frac{\pi}{2} \sum_{\kappa\kappa'} V_{\kappa\kappa\kappa'}^2 [(\bar{n}_{\kappa} + \bar{n}_{\kappa'} + 1) \delta(\omega_{\kappa} - \omega_{\kappa} - \omega_{\kappa'}) + 2(\bar{n}_{\kappa} - \bar{n}_{\kappa'}) \delta(\omega_{\kappa} - \omega_{\kappa} + \omega_{\kappa'})],$$

$$P_{\kappa'} = P \sum_{\kappa\kappa'} V_{\kappa\kappa\kappa'}^2 \left[\frac{(\bar{n}_{\kappa} + \bar{n}_{\kappa'} + 1) (\omega_{\kappa} + \omega_{\kappa'})}{\omega_{\kappa}^2 - (\omega_{\kappa} + \omega_{\kappa'})^2} + \frac{\bar{n}_{\kappa} - \bar{n}_{\kappa'}}{\omega_{\kappa} - \omega_{\kappa} + \omega_{\kappa'}} \right]. \quad (20)$$

4. INVESTIGATION OF SPECTRAL DISTRIBUTION OF DETACHED OSCILLATIONS

It is seen from (4), (18), and (19) that the time-dependent correlation function $\bar{Q}_K(t)$ is expressed in the case considered here in terms of elementary functions, i.e., it can be determined in explicit form for arbitrary relations between $V_{K\kappa\kappa'}$ and $\Gamma_{\kappa'}$. As already noted, the spectra for the absorption or scattering of waves by detached oscillations are determined by Fourier components of this function $\bar{Q}_K(\omega)$. According to (1), (4), (18), and (19), $\bar{Q}_K(\omega)$ can be expressed in

terms of quadratures. For any specified set of parameters, this spectral representation can be determined easily by numerically integrating with respect to t , as was done for the classical limit in [7]. Unlike the classical problem, however, the spectral distribution obtained can now have a fine structure.

The integral that determines $\bar{Q}_K(\omega)$ can be calculated analytically in a number of different limiting cases. We consider first the case of strong nonlinearity, when

$$|V_{\kappa\kappa'}| \gg \Gamma_{\kappa} (1 + 2\bar{n}_{\kappa})$$

and the spectral distribution has a pronounced fine structure. Expanding $a_{K\kappa'}$ in (19) in powers of $\Gamma_{\kappa'}/V_{K\kappa'}$ and retaining the quadratic terms of the expansion, and also expanding

$$\Psi_{\kappa\kappa'}^{-1}(t) \exp(a_{\kappa\kappa'} t) \text{ and } \Psi_{\kappa\kappa'}^{-2}(t) \exp(2a_{\kappa\kappa'} t)$$

in infinite series in $\exp(-a_{K\kappa'} t)$, we express $\bar{Q}_K(t)$ in the form

$$\bar{Q}_{\kappa}(t) = \frac{1}{Z_i} \left(1 - 4i \frac{\Gamma_{\kappa}}{V_{\kappa\kappa}} \bar{n}_{\kappa} \right) \prod_{\kappa'} \left\{ \left(1 - 4i \frac{\Gamma_{\kappa'}}{V_{\kappa\kappa'}} \bar{n}_{\kappa'} \right) \sum_{m_{\kappa'}=1}^{\infty} m_{\kappa'} \exp \left[\Gamma_{\kappa'} t - iP_{\kappa'} \delta_{\kappa\kappa'} t - iV_{\kappa\kappa'} m_{\kappa'} t - \Gamma_{\kappa'} (1 + 2m_{\kappa'} - \delta_{\kappa\kappa'}) \left[1 + 2\bar{n}_{\kappa'} + 4i \frac{\Gamma_{\kappa'}}{V_{\kappa\kappa'}} \bar{n}_{\kappa'} (\bar{n}_{\kappa'} + 1) \right] t - \lambda_{\omega_{\kappa'} m_{\kappa'}} + 4i \frac{\Gamma_{\kappa'}}{V_{\kappa\kappa'}} (m_{\kappa'} - \delta_{\kappa\kappa'}) \right] \right\}. \quad (21)$$

If there is only one detached oscillation or if it is possible to neglect the interaction between the considered oscillation and the others, then all that remains in the product of (18) is a single factor with $\kappa' = \kappa$. Substituting expression (21) in (1) for this case, we obtain a spectral distribution that consists of a set of narrow, almost equidistant lines:

$$Q_{\kappa}(\omega) = \frac{1}{\pi Z_i} \sum_{m=1}^{\infty} m \exp[-\lambda_{\omega_{\kappa}}(m-1)] \frac{\Gamma_{\kappa}(m) \cos \alpha_{\kappa}(m) - \Omega_{\kappa}(m) \sin \alpha_{\kappa}(m)}{\Gamma_{\kappa}^2(m) + \Omega_{\kappa}^2(m)},$$

$$\Omega_{\kappa}(m) = \omega - \omega_{\kappa} - P_{\kappa} - V_{\kappa\kappa} \left(m - \frac{1}{2} \right) - 8 \frac{\Gamma_{\kappa}^2}{V_{\kappa\kappa}} m \bar{n}_{\kappa} (\bar{n}_{\kappa} + 1),$$

$$\Gamma_{\kappa}(m) = \Gamma_{\kappa} [2m(1 + 2\bar{n}_{\kappa}) - 1], \quad \alpha_{\kappa}(m) = 4 \frac{\Gamma_{\kappa}}{V_{\kappa\kappa}} (m - 1 - 2\bar{n}_{\kappa}). \quad (22)$$

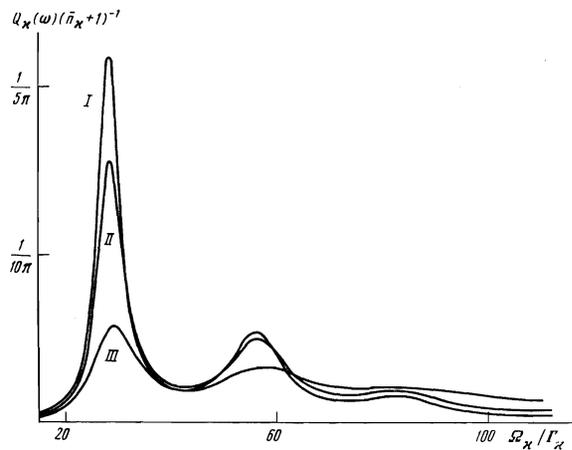
In the limiting case $\Gamma_{\kappa}/V_{K\kappa} \rightarrow 0$ we have

$$\cos \alpha_{\kappa}(m) \approx 1, \quad \sin \alpha_{\kappa}(m) \rightarrow 0,$$

i.e., the individual lines have Lorentz shapes with widths $\Gamma_{\kappa}(m)$. At noticeable values of the ratio $\Gamma_{\kappa}/V_{K\kappa}$, and also at large values of m , it is necessary to take into account also the term $\Omega_{\kappa}(m) \sin \alpha_{\kappa}(m)$ in the numerator of (22), so that the lines become somewhat asymmetrical (and the more so the larger $\alpha_{\kappa}(m)$), and their maxima shift somewhat relative to the points $\Omega_{\kappa}(m) = 0$. We note that the terms discarded in the derivation of (22) give rise to corrections of order $\Gamma_{\kappa}^3 m/V_{K\kappa}^2$ in $\Gamma_{\kappa}(m)$, and $\Gamma_{\kappa}^2 m/V_{K\kappa}^2$ in $\alpha_{\kappa}(m)$.

The widths of the individual small bands increase linearly with increasing line number m and increase with rising temperature, particularly as a result of the factor $2\bar{n}_{\kappa} + 1$.

Although the fine-structure line widths are proportional to the usually defined decay width Γ_{κ} of the detached oscillation, they can in no way be reduced to it. It is easy to verify that these widths are equal to the sums of the reciprocal lifetimes of the upper and lower levels between which the transition corresponding to the considered line takes place. This result agrees with the Weisskopf and Wigner general theory of the widths of spectral lines [8].



Fine structure of spectral distribution $Q_K(\omega) (\bar{n}_K + 1)^{-1}$ for $V_{KK} = 28\Gamma_K$ at different temperatures. On the abscissas we have $\Omega_K = \omega - \omega_K - P_K + V_{KK}/2$. Curve I corresponds to $\bar{n}_K = 3/8$ and $kT = 0.77 \omega_K$, curve II to $\bar{n}_K = 1/2$ and $kT = 0.91 \omega_K$, and curve III to $\bar{n}_K = 1$ and $kT = 1.44 \omega_K$. With increasing temperature, the contribution of the maxima of higher orders increases and the intensity oscillations decrease.

At sufficiently large m , the widths $\Gamma_K(m)$ are comparable to the distance between the lines $|V_{KK}|$, and the narrow bands should overlap. At very low temperatures ($kT \ll \omega_K$) the spectrum consists in practice of only one line (corresponding to $m = 1$ in formula (22)). At somewhat higher temperatures, there appear at first very weak lines corresponding to $m = 2, 3, \dots$. With further rise in temperature, an increase takes place, on the one hand, in the number of narrow bands of noticeable intensity with large m , and on the other hand, in their widths. Therefore the fine structure becomes smeared out to an ever-increasing degree and will gradually vanish, starting with the violet end (at $V_{KK} > 0$) or the red end (at $V_{KK} < 0$) of the spectrum.

At sufficiently high temperatures, such a structure can vanish completely. We note that in some cases the fine structure will thus appear only in a limited interval of intermediate temperatures, and to observe it it is necessary to perform experiments at such temperatures.

To illustrate the vanishing of the fine structure with temperature, the figure shows the spectral distribution curves for a definite set of parameters at different temperatures.

In the case of several interacting detached oscillations at $|V_{KK'}| \gg \Gamma_K' (1 + 2\bar{n}_K)$, it follows from (1) and (21) that $Q_K(\omega)$ is given by

$$Q_K(\omega) = \frac{\exp(\omega_K \lambda)}{\pi Z} \sum_{m_{K'}=1, \dots} m_{K'} \exp\left(-\lambda \sum_{K'} \omega_{K'} m_{K'}\right) \times \frac{\Gamma_K(m_{K'}) \cos \alpha_K(m_{K'}) - \Omega_K(m_{K'}) \sin \alpha_K(m_{K'})}{\Gamma_K^2(m_{K'}) + \Omega_K^2(m_{K'})}, \quad (23)$$

$$\Omega_K(m_{K'}) = \omega - \omega_K - P_K + \frac{V_{KK'}}{2} - \sum_{K_1} V_{KK_1} m_{K_1} - 4 \sum_{K_1} \frac{\Gamma_{K_1}^2}{V_{KK_1}} \bar{n}_{K_1} (\bar{n}_{K_1} + 1) (1 + 2m_{K_1} - \delta_{KK_1}),$$

$$\Gamma_K(m_{K'}) = \sum_{K_1} \Gamma_{K_1} [(1 + 2m_{K_1} - \delta_{KK_1}) (1 + 2\bar{n}_{K_1}) - 1]. \quad (24)$$

$$\alpha_K(m_{K'}) = 4 \sum_{K_1} \frac{\Gamma_{K_1}}{V_{KK_1}} [m_{K_1} - \delta_{KK_1} - \bar{n}_{K_1} (1 + \delta_{KK_1})].$$

In this case each of the lines with $m_K = 1, 2, \dots$ gives rise to a series of lines with $m_{K'} = 0, 1, 2, \dots$, which

are located at a distance $V_{KK'}$ away. The line widths are determined by the values of Γ_{K_1} of all the detached oscillations, and depend linearly on m_{K_1} and \bar{n}_{K_1} . We note that if $|V_{KK'}| \gg \Gamma_{K'}$ for certain detached oscillations, then such oscillations drop out of the sums (24).

In the opposite limiting case when $|V_{KK'}| \ll \Gamma_{K'}$, the fine structure of the spectrum vanishes and the spectral distribution turns out to be smooth. Expanding $a_{KK'}$ and $\Psi_{KK'}(t)$ in (18) and (19) in powers of $|V_{KK'}|/\Gamma_{K'}$ up to terms of second order inclusive, substituting the result of the expansion in (1), we obtain a single smooth distribution

$$Q_K(\omega) = \frac{1}{\pi} (\bar{n}_K + 1) \left\{ \frac{\Gamma_K}{\Gamma_K^2 + \Omega_K^2} + \sum_{K'} \frac{V_{KK'}}{4\Gamma_{K'}^2} \bar{n}_{K'} (\bar{n}_{K'} + 1) (1 + \delta_{KK'}) \left[\frac{\Gamma_{K'}}{\Gamma_{K'}^2 + \Omega_{K'}^2} - \frac{\Gamma_K + 2\Gamma_{K'}}{(\Gamma_K + 2\Gamma_{K'})^2 + \Omega_K^2} - \frac{\Gamma_{K'} (\Gamma_K^2 - \Omega_K^2)}{(\Gamma_K^2 + \Omega_K^2)^2} \right] \right\}$$

$$\Omega_K = \omega - \omega_K - P_K - \frac{V_{KK'}}{2} - \sum_{K'} V_{KK'} \bar{n}_{K'} (1 + \delta_{KK'}). \quad (25)$$

The first term corresponds here to the usual Lorentz distribution with width $2\Gamma_K$, which is determined by the finite lifetime $\tau_K = \Gamma_K^{-1}$ of the detached oscillation. The second correction term has a non-Lorentz shape. The corresponding contribution to the broadening is not determined directly by the finite lifetime and has a modulation character. We note that formula (25) for the limiting case of small $|V_{KK'}|$ agrees with the analogous formula derived in^[15] by the Green's-function method (the formulas in^[5] differ in form from (25), but can be reduced to the same form when account is taken of the condition $|V_{KK'}| \ll \Gamma_{K'}$).

At high temperatures $kT \gg \omega_K$ and at $|V_{KK'}| < \Gamma_{K'} kT/\omega_K$, when there is no fine structure, one can go to the classical limit and obtain a general formula for $Q_K(\omega)$, valid for both large and small ratios $|V_{KK'}|/\Gamma_{K'}$. To this end we take into account the fact that actually $V_{KK'}$ is proportional to the Planck constant \hbar ($V_{KK'} = \hbar \gamma_{kk'} (1 - \delta_{KK'}/4)/\omega_K \omega_{K'}$, where $\gamma_{kk'}$ are the classical nonlinearity constants introduced in^[7]), and, assuming

$$|V_{KK'}| \ll \Gamma_{K'} kT / \omega_{K'},$$

we neglect the last term in formula (19) for $a_{KK'}^2$. Then formulas (1), (18), and (19) lead to the same spectral distribution that was obtained in^[7] by a significantly different method within the framework of the classical theory and was investigated in detail there. The width of the resultant distribution can in this case greatly exceed $\Gamma_K = \tau_K^{-1}$.

In the formulas presented above, the damping $\Gamma_K = \tau_K^{-1}$, which is connected with terms of the type $a_K a_K^+$ or $a_{K'} a_{K'}^+$ in H_i , is due to the finite lifetime of the detached oscillation. In the case of high-frequency detached oscillations, expressions (9) and (20) for the Γ_K connected with these interactions vanish or are small, and the principal role can be assumed by damping due to the interaction

$$H_i = \frac{1}{2} \sum_{KK'} V_{KK'} (a_K + a_{K'}^+) (a_K + a_{K'}^+),$$

which is not connected with the finite lifetime and which has a modulation character (we assume here that $| \omega_K - \omega_{K'} | > 2\omega_m$ at $K \neq K'$, and therefore disregard terms of the type $a_K a_{K'}^+ a_{K'} a_K^+$ with $K \neq K'$). According to (7), these terms introduce in $F^{(1)}(m_{K'}, t)$ an increment of the form

$$-P_{\kappa\kappa'}it - \Gamma_{\kappa\kappa}t - iP_{\kappa\kappa}''(m_{\kappa})t,$$

$$P_{\kappa\kappa}' = \sum_k V_{\kappa\kappa k} (2\bar{n}_k + 1), \quad \Gamma_{\kappa\kappa} = 4\pi \sum_{k,k'} V_{\kappa\kappa k}^2 \bar{n}_k (\bar{n}_k + 1) \delta(\omega_k - \omega_{k'}),$$

and $P_{\kappa\kappa}'' \sim \sum_{k,k'} V_{\kappa\kappa k}^2 V_{\kappa\kappa k'}^2 / \omega_m$ leads to a small correction in comparison with the third-order anharmonicity, and will be disregarded.

By arguments similar to those given above we find that the considered interaction leads to a renormalization of the frequency ω_K by $P_{\kappa\kappa}'$ and to the appearance of an additional factor $\exp(-\Gamma_{\kappa\kappa}t)$ in expression (18) for $\bar{Q}_K(t)$. This means that the resultant spectral distribution is a convolution of the distribution $Q_K(\omega)$, investigated above, and a Lorentz distribution with width $2\Gamma_{\kappa\kappa}$. Accordingly, $\Gamma_K(m_{\kappa\kappa}')$ in formulas (22) and (23) is increased by $\Gamma_{\kappa\kappa}$.

The allowance for an interaction that is nonlinear in the coordinates of the detached oscillations (of the type $a_{\kappa}a_{\kappa'}a_{\kappa}^+$ or $a_{\kappa}a_{\kappa'}a_{\kappa}^+$) is a more difficult special problem, since the partial differential equation of the type (15) contains second derivatives. We therefore do not consider this interaction here in the general case. If, however, we confine ourselves to the case when $\omega_K + \omega_{K'} > \omega_m$ and $|\omega_K - \omega_{K'}| > \omega_m$ at $K \neq K'$, then we can easily verify that an interaction of the type $a_{\kappa}a_{\kappa'}a_{\kappa}^+$ leads only to a renormalization of the non-linearity parameters $V_{\kappa\kappa\kappa}'$ and to a shift of P_K in $\bar{Q}_K(t)$ ($\omega_K - V_{\kappa\kappa\kappa}'/2$ in formula (4), which connects $\bar{Q}_K(t)$ with $Q_K(t)$, is not renormalized):

$$V_{\kappa\kappa\kappa}' \rightarrow V_{\kappa\kappa\kappa}' + 4 \left(1 - \frac{1}{2} \delta_{\kappa\kappa'}\right) \sum_k \left\{ V_{\kappa\kappa\kappa}'^2 \left[\frac{1}{\omega_{\kappa} + \omega_{\kappa'} - \omega_k} - \frac{1}{\omega_{\kappa} + \omega_{\kappa'} + \omega_k} + \frac{1}{\omega_{\kappa'} - \omega_{\kappa} - \omega_k} + \frac{1}{\omega_{\kappa} - \omega_{\kappa'} - \omega_k} \right] - \frac{2V_{\kappa\kappa\kappa}' V_{\kappa'\kappa\kappa}'}{\omega_k} \right\}$$

$$P_{\kappa\kappa} \rightarrow P_{\kappa\kappa} + 4 \sum_{k,k'} \left\{ V_{\kappa\kappa k}^2 \left[\frac{2\bar{n}_k(\omega_k - \omega_{\kappa'})}{(\omega_k - \omega_{\kappa'})^2 - \omega_{\kappa'}^2} - \frac{2(\bar{n}_k + 1)(\omega_k + \omega_{\kappa'})}{(\omega_k + \omega_{\kappa'})^2 - \omega_{\kappa'}^2} \right] \right. \quad (26)$$

$$\left. - \frac{V_{\kappa\kappa k} V_{\kappa'\kappa k}}{\omega_k} \right\} + 4 \sum_k \frac{V_{\kappa\kappa k}^2 (8\omega_k^2 - 3\omega_{\kappa'}^2)}{4\omega_k\omega_{\kappa'}^2 - \omega_{\kappa'}^4},$$

and the foregoing investigation of the shape of the spectral distribution remains applicable.

APPENDIX

The system of ordinary differential equations that determine the characteristics of the partial differential equation (15) is of the form

$$\frac{dt}{ds} = 1, \quad \frac{dx_{\kappa_1}}{ds} = \mu_{\kappa_1}, \quad \frac{dF}{ds} = c_{\kappa} F, \quad (A.1)$$

where s is a parameter that varies along the characteristic curve; μ_{κ_1} and c_{κ} are defined by formulas (17). According to the initial condition (16), the Cauchy prob-

lem corresponds to the determination of an integral surface passing through a line, on which

$$t(0) = 0, \quad x_{\kappa_1}(0) = x_{\kappa_1 0}, \quad F(0) = F(x_{\kappa_1 0}, 0) \quad (A.2)$$

(the $x_{\kappa_1 0}$ describe the line in parametric form).

To solve the system (A.1) with non-separable variables, it is convenient to introduce the change of variable

$$x_{\kappa_1} = iV_{\kappa\kappa}t + \omega_{\kappa_1}\lambda + y_{\kappa_1}. \quad (A.3)$$

Following this substitution, we obtain a system with separable variables y_{κ_1} . Solving this system, we obtain in implicit form the dependence of y_{κ_1} on s and on $y_{\kappa_1 0} = x_{\kappa_1 0} - \omega_{\kappa_1}\lambda$:

$$\frac{\beta_{\kappa\kappa_1} \operatorname{th}(y_{\kappa_1}/2) + \Gamma_{\kappa_1} - a_{\kappa\kappa_1}}{\beta_{\kappa\kappa_1} \operatorname{th}(y_{\kappa_1}/2) + \Gamma_{\kappa_1} + a_{\kappa\kappa_1}} = \frac{\beta_{\kappa\kappa_1} \operatorname{th}(y_{\kappa_1 0}/2) + \Gamma_{\kappa_1} - a_{\kappa\kappa_1}}{\beta_{\kappa\kappa_1} \operatorname{th}(y_{\kappa_1 0}/2) + \Gamma_{\kappa_1} + a_{\kappa\kappa_1}} \exp(2a_{\kappa\kappa_1}s), \quad (A.4)$$

where $\beta_{\kappa\kappa_1} = 2\Gamma_{\kappa_1}(2\bar{n}_{\kappa_1} + 1) + (\frac{1}{2})iV_{\kappa\kappa_1}$, and $a_{\kappa\kappa_1}$ is given by (19).

With the aid of (A.1), (A.2), and (A.4), we can express F in explicit form as a function of s and of the parameters $y_{\kappa_1 0}$. To determine the correlation function $Q_K(t)$, as already noted, it is necessary to know F at $x_{\kappa_1} = \omega_{\kappa_1}\lambda + iV_{\kappa\kappa_1}t$, i.e., at $y_{\kappa_1} = 0$. Recognizing that $s = t$ according to (A.1) and (A.2), the condition $y_{\kappa_1}(s = t) = 0$ and formula (A.4) determine $y_{\kappa_1 0}$ as a function of t . Substituting these values of $y_{\kappa_1 0}(t)$ and $s = t$ into the obtained expression for F , we obtain the function $F(x_{\kappa_1}' = \omega_{\kappa_1}'\lambda + iV_{\kappa\kappa_1}'t, t) = Z_1 \bar{Q}_K(t)$, given in formula (18).

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