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## Spectral Distribution of Nonlinear Oscillators with Nonlinear Friction Due to a Medium

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The analog of a quantum kinetic equation for the description of time correlation functions of singled out nonlinear oscillators interacting with a medium (e.g. local or quasi-local vibrations in crystals) is obtained. Both the frictions, the linear and nonlinear one (the latter is due to decays with participation of two quanta of singled out oscillators), were taken into account. Spectral distributions of the correlation functions of the coordinates are found as well as those of occupation numbers. The essentially non-Lorentzian shape of the spectral distribution and its dependence on interaction parameters and temperature are analysed.

Получен аналог квантового кинетического уравнения для описания временных корреляционных функций выделенных нелинейных осцилляторов, взаимодействующих со средой, например локальных или квазилокальных колебаний в кристаллах. Учитывается как линейное, так и нелинейное трение (обусловленное процессами с участием двух квантов выделенных колебаний). Найдены спектральные распределения корреляторов координат, а также чисел заполнения. Проанализирована существенно нелоренцовская форма спектральных распределений, ее зависимость от параметров взаимодействия и от температуры.

### 1. Introduction

There are some applications where it proves to be necessary to consider the relaxation of singled out nonlinear oscillators interacting with a medium and to calculate the corresponding time correlation functions as well as their spectral distributions. Thus, for instance, local and quasi-local vibrations near defects in crystals are to a more or less degree nonlinear while their spectral distributions determine impurity spectra of infrared absorption and of light or neutron scattering. Some problems connected with nonlinear oscillations arise in the theory of lasers and plasma. The problem of nonlinear oscillator damping is of interest also for the general theory of relaxation in the statistical physics.

Relaxation of linear oscillators with interaction with a medium depending linearly on their coordinates  $q_{\kappa}$  (or the operators  $a_{\kappa}$ ,  $a_{\kappa}^{\dagger}$ ) was investigated in detail in papers [1 to 5]. It is found in particular that such interaction gives rise to an effective friction proportional to the velocity. In this case the spectral representation  $Q_{\kappa}(\omega)$  of the time correlation function  $Q_{\kappa}(t) = \langle q_{\kappa}(t) q_{\kappa}(0) \rangle$  has the well-known Lorentzian shape whose half-width is equal to the double reciprocal lifetime  $2\tau_{\kappa}^{-1}$  of the oscillation  $\kappa$ . As it is shown in the [6] even small nonlinearity of singled out oscillators may result in a significant modi-

fication of the shape of the distribution  $Q_{\kappa}(\omega)$  and may produce an essential additional broadening which is not connected with the lifetime and has a modulational nature. In terms of the classical theory these effects are due to the dependence of the nonlinear oscillation frequency on the amplitude that is to the frequency modulation in time (as a consequence of interaction with a medium) while in terms of the quantum theory they are due to the non-equidistance of levels of the nonlinear oscillator. With the use of special methods developed in the classical [7] and quantum [8] theory the complete calculation of  $Q_{\kappa}(t)$  was performed for nonlinear oscillators with a linear friction (the energy of interaction with a medium is linear in  $q_{\kappa}$ ).

New effects arise in the case of nonlinear friction i.e. when the interaction energy contains terms proportional to  $q_{\kappa}^2$ . Since here the friction coefficient depends on the amplitude and hence varies in time even for a linear (with neglect of interaction) oscillator the relaxation process becomes complex, the damping of the correlator  $Q_{\kappa}(t)$  will be non-exponential and the spectral distribution  $Q_{\kappa}(\omega)$  will be non-Lorentzian.  $Q_{\kappa}(\omega)$  becomes still more complex if both the restoring force and the friction are nonlinear. The friction nonlinearity especially strongly affects the correlation function of the occupation number operators  $N_{\kappa}(t) = \langle \hat{n}_{\kappa}(t) (\hat{n}_{\kappa}(0) - \bar{n}_{\kappa}) \rangle$  ( $\hat{n}_{\kappa} = a_{\kappa}^{\dagger} a_{\kappa}$ ,  $\bar{n}_{\kappa} = \langle \hat{n}_{\kappa} \rangle$ ). In case of linear friction modulational effects do not influence this function and it decreases as  $\exp(-2t/\tau_{\kappa})$  even for a nonlinear oscillator. But if the friction is not linear the law of damping becomes essentially nonexponential.

Below the quantum theory of singled out oscillators with nonlinear friction is developed with the aid of the method proposed in the earlier paper [8]. In Section 2 the general equation is obtained which is the analog of a quantum-kinetic equation for the time correlation function of arbitrary operators of such oscillators. This equation is then used for the analysis of the spectral distribution of a correlator for the coordinates of a single isolated oscillation (Section 3) or two interacting oscillations (Section 4). Numerical calculations allow to investigate the distribution shape for various parameter values. In Section 5 the correlator of the occupation numbers is analysed in the similar way.

## 2. Quantum Kinetic Equation for the Time Correlation Function

For the sake of definition we shall consider here as singled out oscillations the oscillations those of a weakly bound impurity atoms or molecules in crystals while as medium we consider an assembly of continuous oscillations in the crystal from which the impurity is taken away. However, the results are practically applicable to general case provided the parameter values in the formulae are changed accordingly. For simplicity we shall confine ourselves to defects of such a symmetry that terms of the third order in  $q_{\kappa}$  are eliminated from the Hamiltonian (this corresponds, e.g., to an impurity atom in the inversion centre).

Singled out oscillations will be denoted by the index  $\kappa$  while oscillations of the continuous spectrum by  $k$ . When there is no interaction the Hamiltonian of the system may be written as

$$H_0 = \sum_{\kappa} \omega_{\kappa} \hat{n}_{\kappa} + \frac{1}{2} \sum_{\kappa \kappa'} V_{\kappa \kappa'} \hat{n}_{\kappa} \hat{n}_{\kappa'} + \sum_k \omega_k \hat{n}_k ;$$

$$\hat{n}_{\kappa} = a_{\kappa}^{\dagger} a_{\kappa}, \quad \hat{n}_k = a_k^{\dagger} a_k. \quad (1)$$

Here  $\omega_\kappa$  and  $\omega_k$  are the frequencies of the oscillations  $\kappa$  and  $k$ ; the second term describes the nonlinearity of the singled out oscillations and their interaction with each other. We have omitted in (1) terms of the type  $a_\kappa^3 a_\kappa^\dagger$  and terms describing the anharmonicity of the oscillations of a continuous spectrum which are of no significance for the effects under consideration. The assumption is made that  $\omega_\kappa$ 's are not close to each other as well as to the combined frequencies  $\omega_{\kappa'} \pm \omega_{\kappa''}$  and do not coincide with singular points in the density of the states of continuous oscillations.

The Hamiltonian of interaction between singled out oscillations and oscillations of the medium when the anharmonicity of fourth order is neglected may be written as

$$H_i = \sum_{\kappa k} V_{\kappa k} (a_\kappa + a_\kappa^\dagger) (a_k + a_k^\dagger) + \frac{1}{2} \sum_{\kappa k k'} V_{\kappa k k'} (a_\kappa + a_\kappa^\dagger) (a_k + a_k^\dagger) (a_{k'} + a_{k'}^\dagger) + \sum_{\kappa \kappa' k} V_{\kappa \kappa' k} (a_\kappa + a_\kappa^\dagger) (a_{\kappa'} + a_{\kappa'}^\dagger) (a_k + a_k^\dagger). \quad (2)$$

The first two terms in this interaction Hamiltonian lead to a linear friction while the last term leads to a nonlinear one.

Let us consider the time correlation function of two arbitrary operators  $A$ ,  $B$  for one of the singled out oscillators  $\kappa$ :

$$\langle A(t) B(0) \rangle = Z^{-1} \text{Sp} (e^{-\lambda H} e^{iHt} A e^{-iHt} B). \quad (3)$$

Here  $H = H_0 + H_i$ ;  $Z = \text{Sp} e^{-\lambda H}$ ;  $\lambda = (kT)^{-1}$ ;  $\hbar = 1$ . Without loss of generality we put  $\langle A \rangle = 0$ . Let us assume also that  $\langle A(0) B(0) \rangle \neq 0$ .

The parameters  $V_{\kappa \kappa'}$  and polarization operators  $R_\kappa$  connected with the interaction  $H_i$  are taken to be small as compared with  $\omega_\kappa$  and  $kT$ . Since, however, we are interested in correlation functions at large times  $t$ ,  $V_{\kappa \kappa'} t$  and  $R_\kappa t$  must not be treated as small ones and the usual perturbation theory fails. Therefore, in calculating the correlators one should use the methods of asymptotic perturbation theory.

We shall confine ourselves at first to a simpler case when there is only one singled out oscillator  $\kappa = 1$  or when the interaction between singled out oscillators may be neglected. The trace in (1) may be calculated using the complete set of eigenfunctions  $(n_\kappa|$  and  $|n_k|$  of harmonic oscillators in the occupation number representation. Let us pass to the interaction representation by introducing the operator  $U(t) = \exp(iH_0 t) \exp(-iHt)$ . Then the expression for the correlator will take the form

$$\langle A(t) B(0) \rangle = (\bar{n} + 1)^{-1} \sum_{m, n, p, q=0}^{\infty} A_{mn} B_{pq} e^{-\lambda \omega_\kappa m} \times \exp\{-i[\omega_\kappa + \frac{1}{2} V(n+m)](n-m)t\} F(n, m; q, p; t). \quad (4)$$

For brevity we have omitted the index  $\kappa = 1$  at the quantum numbers  $m, n, p, q$ , the mean Planck number  $\bar{n} \equiv \bar{n}_1$  and the nonlinearity constant  $V_{11} \equiv V$ ;  $A_{mn}$  and  $B_{pq}$  denote the matrix elements of the operators  $A$  and  $B$  with respect to the harmonic oscillator wave functions while the function  $F$  is defined by the formula

$$F(n, m; q, p; t) = \prod_k (\bar{n}_k + 1)^{-1} \sum_{\dots, n_k, m_k, \dots=0}^{\infty} \exp\left(-\lambda \sum_k \omega_k n_k\right) \times \langle n, n_k | U(t) | p, m_k \rangle \langle q, m_k | U^{-1}(t + i\lambda) | m, n_k \rangle. \quad (5)$$

Using the chronological ordering of operators and taking into account the smallness of the coefficients  $V_{\alpha k k'}$  one can carry out the summation over  $n_k, m_k$  in (5) that excludes the continuous oscillations. Then the procedure similar to that used in [8] makes it possible to deduce the differential-difference equation for the function  $F$  valid (in the sense of the asymptotic perturbation theory) in the large time range  $t \gg \omega_\alpha^{-1}, \omega_m^{-1}$  ( $\omega_m$  is the maximum frequency of the continuous spectrum). This equation plays the role of a quantum kinetic equation and has the form

$$\frac{\partial F(n, m; q, p; t)}{\partial t} = \sum_{j=-2}^2 D_j(n, m) e^{-iV_j(n-m)t} F(n+j, m+j; q, p; t), \quad (6)$$

where the coefficients  $D_j$  are defined by the expressions

$$\left. \begin{aligned} D_0(n, m) &= -\Gamma(n) - \Gamma(m) - iP(n-m) - i\frac{V^{(2)}}{2}(n-m)(n+m+1); \\ D_{\pm 1}(n, m) &= 2\Gamma(\bar{n} + \frac{1}{2} \mp \frac{1}{2}) \sqrt{(n + \frac{1}{2} \pm \frac{1}{2})(m + \frac{1}{2} \pm \frac{1}{2})}; \\ D_{\pm 2}(n, m) &= 2\Gamma^{(2)}(\bar{n}_2 + \frac{1}{2} \mp \frac{1}{2}) \sqrt{(n \pm 1)(n+1 \pm 1)(m \pm 1)(m+1 \pm 1)}. \end{aligned} \right\} \quad (7)$$

Here  $P$  is the shift of the frequency of the singled-out oscillator due to interaction with the medium. It is determined by equations (9), (20), (26) for  $P_\alpha$  in [8].  $\tilde{V}^{(2)}$  is the renormalization of the non-equidistancy constant  $V$  defined by the second addend in equation (26) in [8] for  $V_{\alpha\alpha}$  (the sums in (26) are to be understood in the sense of a principal value). The parameters  $\Gamma$  are related with the damping of the singled out oscillation:

$$\left. \begin{aligned} \Gamma(m) &= \Gamma[m(2\bar{n}+1) + \bar{n}] + \Gamma^{(2)}[2\bar{n}_2(m^2 + m + 1) + m(m-1)]; \\ \bar{n}_2 &= \bar{n}(2\omega_\alpha) = \frac{\bar{n}^2}{2\bar{n}+1}. \end{aligned} \right\} \quad (8)$$

$$\begin{aligned} \Gamma &= \pi \sum_k V_{\alpha k}^2 \delta(\omega_\alpha - \omega_k) + \frac{\pi}{2} \sum_{kk'} V_{\alpha k k'}^2 [(\bar{n}_k + \bar{n}_{k'} + 1) \delta(\omega_\alpha - \omega_k - \omega_{k'}) + \\ &\quad + 2(\bar{n}_{k'} - \bar{n}_k) \delta(\omega_\alpha - \omega_k + \omega_{k'})]; \quad \bar{n}_k = \langle \hat{n}_k \rangle. \end{aligned} \quad (9)$$

$$\Gamma^{(2)} = \pi \sum_k V_{\alpha k}^2 \delta(2\omega_\alpha - \omega_k). \quad (10)$$

The coefficients  $D$  in equation (6) as well as in [8] were calculated to the second approximation of the perturbation theory. The initial condition for this equation is evident  $F(n, m; q, p; 0) = \delta_{n,p} \delta_{m,q}$ . Since the solutions of equation (6) corresponding to  $m, n \geq 0$  and to  $m, n < 0$  are not connected this equation and the initial condition unambiguously determine  $F$  at all  $t$ .

In order to calculate correlation functions from (4), (6) in any concrete case it is convenient to multiply (6) by  $B_{pq}$  and to perform the summation over  $p, q$  making use of the fact that the factors in (6) do not depend on  $p, q$ . Then for the function

$$F_B(n, m; t) = \sum_{p,q} F(n, m; q, p; t) B_{pq} \quad (F_B(n, m; 0) = B_{nm}) \quad (11)$$

we get the same equation (6) with the initial condition written in (11).

### 3. The Spectral Distribution of the Time Correlation Function of Oscillator Coordinates

The shape of lines in spectra of infrared absorption or emission (or inelastic neutron scattering) by local or quasi-local vibrations is determined by the spectral representation  $Q_{\kappa}(\omega) = \langle a_{\kappa}(t) a_{\kappa}^{\dagger}(0) \rangle_{\omega}$  with an accuracy to a factor weakly depending on frequency. Equations (4), (11) being taken into account,  $Q_{\kappa}(\omega)$  may be written in the form

$$\left. \begin{aligned} Q_{\kappa}(\omega) &= \frac{1}{\pi} (\bar{n} + 1)^{-1} \operatorname{Re} \sum_{m=1}^{\infty} \varphi(m, \omega); \\ \varphi(m, \omega) &= \int_0^{\infty} dt \exp(i\Omega t) \sum_{q=1}^{\infty} F(m, m-1; q-1, q; t) \sqrt{mq} \times \\ &\quad \times \exp[-\lambda\omega_{\kappa}(m-1) - iVmt]; \quad \Omega = \omega - \omega_{\kappa} - P + \frac{1}{2} V. \end{aligned} \right\} \quad (12)$$

The functions  $\varphi(m, \omega)$  with different  $m$  satisfy the difference equation which follows from (6) written for the function (11) with  $B = a_{\kappa}^{\dagger}$  after the transition to the spectral representation in (6) and taking into account the expressions for the matrix elements of the operator  $A = a_{\kappa}$

$$\left. \begin{aligned} [i(\Omega - \tilde{V}m) - \Gamma(m) - \Gamma(m-1)] \varphi(m, \omega) + \sum_{j=0} D_j(m, m-1) e^{j\omega_{\kappa}} \times \\ \times \sqrt{\frac{m}{m+j}} \varphi(m+j, \omega) = -m \exp[-\lambda\omega_{\kappa}(m-1)]; \quad \tilde{V} = V + V^{(2)}; \\ \varphi(0, \omega) = \varphi(\infty, \omega) = 0. \end{aligned} \right\} \quad (13)$$

Each addend  $\varphi(m, \omega)$  in the sum (12) determines the partial spectrum corresponding to the transition from the level  $m-1$  to the  $m$ -th one. It is essential, however, that these transitions "interfere" and are not independent. In fact, if to neglect the terms with  $j \neq 0$  equation (13) describes the Lorentzian shape of the line  $\operatorname{Re} \varphi(m, \omega)$  for a single transition, but the terms with  $j \neq 0$  complicate in a general case the shapes of both the individual lines and the whole distribution.

When  $\Gamma$  and  $\Gamma^{(2)}$  (and hence  $D_j$  ( $j \neq 0$ )) are small one can solve (13) by the iteration method neglecting in the zeroth approximation the terms with  $j \neq 0$  at the determination of  $\varphi = \varphi^0$  but allowing for these terms in the first approximation (with  $\varphi$  being replaced by  $\varphi^0$ ). Thus

$$\left. \begin{aligned} \operatorname{Re} \varphi(m, \omega) &= m \exp[-\lambda\omega_{\kappa}(m-1)] \Phi(m, \omega) \times \\ \times \left[ 1 + \sum_{j=\pm 1, \pm 2} D_j(m, m-1) \sqrt{\frac{m+j}{m}} \Phi(m+j, \omega) \frac{\Gamma_m \Gamma_{m+j} - \Omega_m \Omega_{m+j}}{\Gamma_m \Gamma_{m+j}} \right]; \\ \Phi(m, \omega) &= \frac{\Gamma_m}{\Gamma_m^2 + \Omega_m^2}; \quad \Gamma_m = \Gamma(m) + \Gamma(m-1); \quad \Omega_m = \Omega - \tilde{V}m. \end{aligned} \right\} \quad (14)$$

The main first term in this expression determines the symmetric Lorentzian curve with maximum at  $\Omega = \tilde{V}m$  that is the lines corresponding to different  $m$  are equidistant in the zeroth approximation. The expression for the half-width of the Lorentzian curve  $2\Gamma_m$  coincides with that obtained according to the Weisskopf-

Wigner formula. Its dependence on the line number  $m$  and on the temperature in case of a linear and nonlinear friction is determined by equations (8) to (10), (14).

The correction term  $\sim \Gamma_m/\tilde{V}$ , appears because of the fact that transition frequencies of the weakly nonlinear oscillator under study are almost degenerate. It leads to some asymmetry in individual separate bands,  $\sim \Gamma_m/\tilde{V}$ , a small non-equidistancy of peaks, and to a change of their intensities. The correcting term is small and the iteration method used here is valid if the condition

$$|\tilde{V}| \gg \Gamma(m) \quad (15)$$

is satisfied. In this case the spectrum has a distinct fine structure. Such a structure was observed in the spectrum of infrared absorption by non-degenerate quasi-local oscillations of impurity ions  $\text{Eu}^{2+}$  in  $\text{MnF}_2$  [9]. According to (14), (8) to (10) the bands are asymmetric, they broaden and shift with temperature increase.

Since  $\varphi(m, \omega)$  is proportional to  $\exp[-\lambda\omega_*(m-1)]$ , only one band corresponding to  $m=1$  appears in the spectrum at  $T \rightarrow 0$ . Other bands are included with temperature rise. If it is possible to ignore all  $\varphi(m, \omega)$  with  $m > 2$  that is to take into account the existence of only two bands then system (13) is reduced to two equations whose solution is of the form

$$\varphi(1, \omega) + \varphi(2, \omega) = \frac{2\bar{n}(\Gamma_1 - i\Omega_1) + \Gamma_2 - i\Omega_2 + 8I\bar{n}}{(\Gamma_1 - i\Omega_1)(\Gamma_2 - i\Omega_2) - 8I^2\bar{n}}. \quad (16)$$

Equation (16) permits to easily calculate the shape of the spectrum  $\text{Re } \varphi(1, \omega) + \text{Re } \varphi(2, \omega)$  for arbitrary parameter values at low temperatures.

This spectrum is composed of the intensive band  $m=1$ , distorted due to interference with the band  $m=2$ , and the band  $m=2$  itself. The spectrum must have two peaks (of essentially different intensities) if  $|\tilde{V}|/\Gamma^{(2)}$  is large enough or must be described by a single asymmetric curve if this ratio is small.

The system (13) has an exact solution in the special case of the complete degeneration of the transition frequencies  $\tilde{V}=0$  and  $\Gamma^{(2)}=0$  (the friction is pure linear). Here

$$\text{Re } \varphi(m, \omega) = m \exp[-\lambda\omega_*(m-1)] \frac{\Gamma}{\Gamma^2 + \Omega^2},$$

that is all the bands are of the same shape and have a common maximum, differing only in intensities. The band width does not depend on the number and naturally does not reduce to the line width calculated according to Weisskopf-Wigner in this case of complete degeneration. The introduction of bands itself is justified here only by their connection with the case of absent degeneration. In such a way, from the adopted point of view the known "paradoxon" of the harmonic oscillator (see, e.g., [3]) may be solved.

The condition (15) ceases to be fulfilled with the increase of temperature or number  $m$ . The fine structure is gradually smeared out (if it was distinct) and equation (14) becomes inapplicable. The opposite extreme case  $|\tilde{V}| \ll \Gamma$  and  $\Gamma^{(2)} \ll \Gamma$  was treated by the Green function method in [6]. It should be noted that the increase of  $T$  and  $m$  makes more important the role of nonlinear friction according to (8) to (10), (13).

It is not difficult also to study analytically the behaviour of the distribution in the wings. Neglecting  $\tilde{V}m$  as compared to  $\Omega$  and noting that the terms with  $j \neq 0$  in (13) are of the order  $\Gamma_m |\varphi| \ll |i\Omega\varphi|$  we get as a result of iteration

$$Q_n(\omega) = \frac{1}{\pi} (\bar{n} + 1) \frac{\Gamma + 4\Gamma^{(2)}(\bar{n} - \bar{n}_2)}{\Omega^2}. \quad (17)$$

The spectral distribution may be analysed in a general case by means of a numerical solution of (13). It is not difficult to solve numerically this system of difference equations and to find both  $\text{Re } \varphi(m, \omega)$  and  $Q_n(\omega)$  for any given set of parameters  $\tilde{V}$ ,  $\Gamma$ ,  $\Gamma^{(2)}$ , and  $T$ . In case of linear friction ( $\Gamma^{(2)} = 0$ ) the solution reduced to is a quadrature of elementary functions. Corresponding numerical calculations for the classical and quantum cases are given in [7, 8]. When the nonlinear friction is also essential the complete set of independent parameters  $\tilde{V}/\Gamma$ ,  $\Gamma^{(2)}/\Gamma$ , and  $T/\omega_n$  is large and therefore we have carry out numerical calculations hereonly for the high temperature range where the fine structure is absent,  $T > \omega_n$  and the classical approximation is applicable.

In the classical limit the main contribution into  $Q_n(\omega)$  give terms with large  $m$ . Let us introduce the continuous variable  $x = \lambda\omega_n m$  and the function  $W(x, \omega) = \lambda\omega_n \varphi(m, \omega)$ . This allows to go from the difference equation (13) to the differential equation

$$\left. \begin{aligned} & 2x(\Gamma + \gamma^{(2)}x) \frac{d^2 W}{dx^2} + 2x[\Gamma + \gamma^{(2)}(1+x)] \frac{dW}{dx} + \\ & + [i(\Omega - vx) + \Gamma - \gamma^{(2)}(1-3x)] W = -xe^{-x}; \quad W(0) = W(\infty) = 0; \\ & \gamma^{(2)} = \frac{2\Gamma^{(2)}}{\lambda\omega_n}; \quad v = \frac{\tilde{V}}{\lambda\omega_n}; \quad Q_n(\omega) = \frac{1}{\pi\lambda\omega_n} \text{Re} \int_0^\infty W(x, \omega) dx. \end{aligned} \right\} (18)$$

The dependences of  $Q_n$  on  $\Omega$  at various  $v/\Gamma$  are plotted in [7] for the case of a purely linear friction ( $\gamma^{(2)} = 0$ ). In the opposite extreme case where nonlinear friction is of the primary importance ( $\Gamma = 0$ ,  $\gamma^{(2)} \neq 0$ ) spectral distributions at various  $v/\gamma^{(2)}$  were found by the numerical solution of the boundary-value problem (18). Fig. 1 shows the results of these calculations (performed on the computer BESM-6). It is seen in this plot that in case of a nonlinear friction even in the absence of non-equidistancy the spectral distribution being symmetrical differs essentially from the Lorentzian one (referred to the case of linear friction). Although curve 1 decreases as  $\gamma^{(2)}/\Omega^2$  at large  $|\Omega|$ , it is significantly narrower and higher in the central part of the spectrum than the Lorentzian curve. The peak height of curve 1 equals  $1.67/\pi$  (instead of  $1/\pi$  for the Lorentzian curve) while the width at the half peak height is half as great as that of the Lorentzian curve. The rise in  $v$  (the parameter  $v$  determines the non-equidistancy of levels) leads to broadening of spectral distributions and to a shifting of their maxima.

In the general case where both the linear and nonlinear friction are present, the problem includes two parameters even at high temperatures and it is inexpedient to give the results of calculations of spectral distributions. Therefore

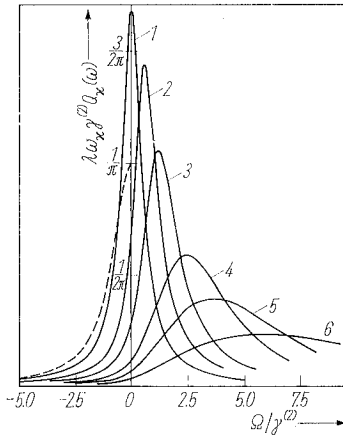


Fig. 1

Fig. 1. Spectral distributions  $Q_n(\omega)$  at high temperatures in the case of purely nonlinear friction. The curves 1 to 6 correspond to the parameter values  $v/\gamma^{(2)} = 0; 0.5; 1; 2; 3; 5$ . The dashed line is relevant to the Lorentzian distribution of the same integral intensity and the same asymptotes at large  $|\Omega|$  as those in the curve 1

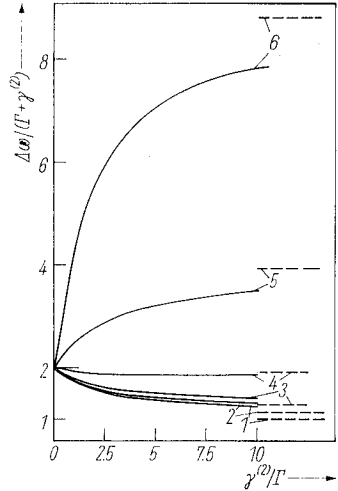


Fig. 2

Fig. 2. The half-widths  $\Delta\omega$  of spectral distributions at various values of the parameters  $v/\gamma^{(2)}$  and  $\gamma^{(2)}/\Gamma$ . The curves 1 to 6 correspond to the parameter values  $v/\gamma^{(2)} = 0; 0.3; 0.5; 1; 2; 4$ . The dashed lines show the asymptotes for the dependences of  $\Delta\omega$  on  $v/\gamma^{(2)}$  at  $\gamma^{(2)}/\Gamma \rightarrow \infty$  (or  $\Gamma \rightarrow 0$ ), corresponding to Fig. 1. The scale is chosen to be proportional to the Lorentzian wing amplitude of the distribution

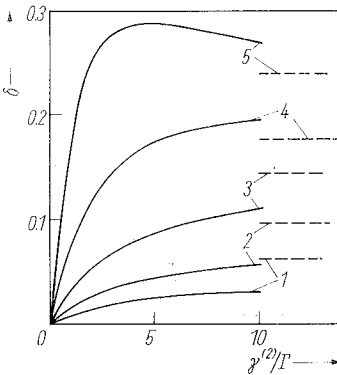


Fig. 3

Fig. 3. Parameters of spectral distribution asymmetry at various values of  $v/\gamma^{(2)}$  and  $\gamma^{(2)}/\Gamma$ . The curves 1 to 5 correspond to  $v/\gamma^{(2)} = 0.3; 0.5; 1; 2; 4$ . The dashed lines show the asymptotics of the dependences of  $\delta$  on  $v/\gamma^{(2)}$  at  $\gamma^{(2)}/\Gamma \rightarrow \infty$

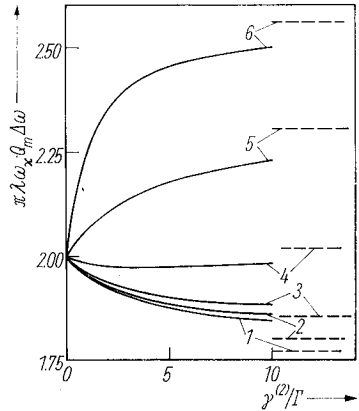


Fig. 4

Fig. 4. Characteristics of a non-Lorentzian shape of the distribution  $Q_m\Delta\omega$  at different values of  $v/\gamma^{(2)}$  and  $\gamma^{(2)}/\Gamma$ . The curves 1 to 6 correspond to  $v/\gamma^{(2)} = 0; 0.3; 0.5; 1; 2; 4$

we shall confine ourselves only to the data for the main characteristics: the widths at a half peak height  $\Delta\omega$ , asymmetry parameters  $\delta = \frac{\Omega_{+1/2} - \Omega_{-1/2}}{\Delta\omega}$  ( $\Omega_{\pm 1/2}$

are the frequency intervals between the points corresponding to the maximum and half peak intensities on the right and left sides of the maximum;  $\Delta\omega = \Omega_{+1/2} + \Omega_{-1/2}$ ), the products  $Q_m\Delta\omega$  ( $Q_m$  is the peak height of the curve).

As can be seen from Fig. 2 the distribution width  $\Delta\omega$  increases with the rise in  $\gamma^{(2)}$  and  $v$  (Fig. 2 to 4 do not show results for  $\gamma^{(2)} = 0$ ,  $v \neq 0$  since they are given in [7]). The distribution asymmetry  $\delta$  shown in Fig. 3 depends monotonously on the parameter  $v$ . The quantity  $\pi\lambda\omega_\kappa Q_m \Delta\omega$  given in Fig. 4 is one of the parameters that describe the departure of the distribution  $Q_\kappa(\omega)$  from the Lorentzian one. It is equal to 2 for the Lorentzian curve. Non-equidistancy causes this parameter to grow while nonlinearity of the friction reduces its value (at  $v/\gamma^{(2)} = 1$  these effects are almost compensated). The dotted lines in Fig. 2 to 4 show asymptotes of the corresponding functions at  $\gamma^{(2)}/\Gamma \rightarrow \infty$  (or  $\Gamma \rightarrow 0$ ). The asymptotes are reached at very large  $\gamma^{(2)}/\Gamma \approx 10^2$ . In the opposite case  $\gamma^{(2)}/\Gamma \lesssim 10^{-1}$  and  $|v|/\Gamma \lesssim 10^{-1}$  the next analytic expressions are valid,

$$\frac{\Delta\omega}{\Gamma + \gamma^{(2)}} = 2 - \frac{26}{15} \frac{(\gamma^{(2)})^2 - v^2}{\Gamma^2}, \quad \delta = \frac{16}{45} \frac{\gamma^{(2)}v}{\Gamma^2},$$

$$\pi\lambda\omega_\kappa Q_m \Delta\omega = 2 - \frac{2}{5} \frac{(\gamma^{(2)})^2 - v^2}{\Gamma^2}.$$

#### 4. The Effect of Interaction between Singled out Oscillators on Their Spectral Distribution

To illustrate the effects due to interaction between singled out oscillators we shall consider the case of two oscillators  $\kappa = 1, 2$ . Carrying out calculations similar to those in the above case of a single oscillator one may write the spectral representation  $Q_1(\omega)$  of the correlator  $\langle a_1(t) a_1^+(0) \rangle$  as the sum of functions of type (12),

$$Q_1(\omega) = \frac{1}{\pi} (\bar{n}(\omega_1) + 1)^{-1} (\bar{n}(\omega_2) + 1)^{-1} \sum_{m_1, m_2=0}^{\infty} \varphi(m_1, m_2, \omega), \quad (19)$$

where  $\varphi$  is determined from difference equation similar to (13),

$$\begin{aligned} & \left[ i \left( \Omega - \sum_{\kappa} \tilde{V}_{1\kappa} m_{\kappa} \right) - \Gamma(m_1, m_2) - \Gamma(m_1 - 1, m_2) \right] \varphi(m_1, m_2, \omega) + \\ & + \sum_{j_1, j_2 = \pm 1, \pm 2} D_{j_1 j_2} \sqrt{\frac{m_1}{m_1 + j_1}} \exp \left( \sum_{\kappa} j_{\kappa} \lambda \omega_{\kappa} \right) \varphi(m_1 + j_1, m_2 + j_2, \omega) = \\ & = - m_1 \exp \left[ - \sum_{\kappa} \lambda \omega_{\kappa} (m_{\kappa} - \delta_{\kappa 1}) \right]. \end{aligned} \quad (20)$$

Here

$$\left. \begin{aligned}
 \Gamma(m_1, m_2) &= \Gamma(m_1) + \Gamma(m_2) + 4\Gamma_{12}^+[m_1 m_2 (\bar{n}_+ + 1) + \\
 &\quad + (m_1 + 1)(m_2 + 1)\bar{n}_+] + 4\Gamma_{12}^- \sum_{\kappa \neq \kappa'} m_{\kappa} (m_{\kappa'} + 1) [\bar{n}_- + \theta(\omega_{\kappa} - \omega_{\kappa'})]; \\
 \bar{n}_{\pm} &= \bar{n}(|\omega_1 \pm \omega_2|); \\
 \Gamma_{12}^{\pm} &= \pi \sum_k V_{12k}^2 \delta(|\omega_1 \pm \omega_2| - \omega_k); \quad \theta(x) = \begin{cases} 1, & x > 0 \\ 0, & x < 0 \end{cases} \\
 D_{j_1 0} &= D_{j_1}(m_1, m_1 - 1); \quad D_{0 j_2} = D_{j_2}(m_2, m_2); \quad D_{\pm 2 \pm 2} = 0; \\
 D_{j_1 j_2} &= 8\Gamma_{12}^+(\bar{n}_+ + \frac{1}{2} - \frac{1}{2} j_1) (m_2 + \frac{1}{2} + \frac{1}{2} j_2) \sqrt{(m_1 + j_1) m_1}, \\
 &\quad (j_1 = j_2 = \pm 1); \\
 D_{j_1 j_2} &= 8\Gamma_{12}^-(\bar{n}_- + \frac{1}{2} - \frac{1}{2} j_1 - j_2 \theta(\omega_2 - \omega_1)) (m_2 + \frac{1}{2} + \frac{1}{2} j_2) \times \\
 &\quad \times \sqrt{m_1(m_1 + j_1)}, \quad (j_1 = -j_2 = \pm 1).
 \end{aligned} \right\} (21)$$

Equation (20) may be solved numerically. A simple analytical solution is obtained for high-frequency local vibrations at  $\lambda\omega_1 \gg 1$  which interact with low-frequency quasi-local vibrations ( $\lambda\omega_2 \geq 1$ ) in case when the conservation law allows the decay of oscillation 1 to the oscillation 2 and the continuous spectrum oscillation  $k$  and when the intrinsic nonlinear friction  $\Gamma^{(2)}$  of the oscillation 2 may be neglected. Then we get the set of differential-difference time-dependent equations corresponding to (20) which is identical with that developed for nonlinear oscillators with a linear friction [8] the only difference being in the explicit form of the constants. Using the results of [8] the following expressions may be obtained for the correlator  $Q_1(t)$  and its spectral representation

$$\left. \begin{aligned}
 Q_1(\omega) &= \frac{1}{\pi} \operatorname{Re} \int_0^{\infty} Q_1(t) \exp(i\omega t) dt; \\
 Q_1(t) &= \exp \left[ -i \left( \omega_1 + P_1 - \frac{1}{2} V_{11} - \frac{1}{2} \tilde{V}_{12} + \tilde{V}_{11} \right) t - \right. \\
 &\quad \left. - (\Gamma_1 - \Gamma_2 + 2\Gamma_{12}^-) t \right] \left\{ \operatorname{ch} at + \left[ 1 + \frac{i\tilde{V}_{12} + 4\Gamma_{12}^-}{2\Gamma_2} (2\bar{n}(\omega_2) + 1) \right] \times \right. \\
 &\quad \left. \times \frac{\Gamma_2}{a} \operatorname{sh} at \right\}^{-1}, \quad t \gg \omega_2^{-1}; \\
 a^2 &= \Gamma_2^2 + \Gamma_2^2 (i\tilde{V}_{12} + 4\Gamma_{12}^-) (2\bar{n}(\omega_2) + 1) + \frac{1}{4} (i\tilde{V}_{12} + 4\Gamma_{12}^-)^2.
 \end{aligned} \right\} (22)$$

Expression (22) describes, generally speaking, the complex asymmetric distribution that may have a fine structure (corresponding to different levels of oscillation 2). This structure is revealed when the oscillation interaction constant  $|\tilde{V}_{12}|$  is large as compared with  $\Gamma_1$ ,  $\Gamma_2$ , and  $\Gamma_{12}^-$ . In this case the separation between bands is  $|\tilde{V}_{12}|$  while the width of the  $m_2$ -th peak is  $\Gamma_{m_2} = \Gamma_1 + \Gamma_2(1 + 2m_2) (1 + 2\bar{n}(\omega_2)) + 4\Gamma_{12}^-(m_2 + 1) - \Gamma_2$ . The main contribution to these widths is connected with the term with  $\Gamma_{12}^-$ .

The fine structure becomes smeared out with decreasing  $|\tilde{V}_{12}|$ . At  $\tilde{V}_{12} = 0$  the spectrum becomes symmetric but its shape is essentially non-Lorentzian. In particular, if  $\tilde{V}_{12} = 0$  and  $\Gamma_{12}^- \gg \Gamma_2$ , we have

$$Q_1(\omega) = \frac{1}{\pi} [\bar{n}(\omega_2) + 1]^{-1} \sum_{m_2=0}^{\infty} \frac{\Gamma_1 + 4\Gamma_{12}^-(m_2 + 1)}{[\Gamma_1 + 4\Gamma_{12}^-(m_2 + 1)]^2 + \Omega^2} \exp(-\lambda\omega_2 m_2). \quad (23)$$

The central part of this distribution is narrower and sharper than the Lorentzian curve possessing the same wings and the same integral intensity.

### 5. The Time Correlation Function of the Occupation Numbers

Nonlinear friction being taken into account the form of the time correlation function  $N_x(t) = \langle \hat{n}_x(t) (\hat{n}_x(0) - \bar{n}_x) \rangle$  is qualitatively changed. To define it in case of only one singled out oscillator one may use equations (4), (6), (7). Then

$$N_1(t) = (\bar{n} + 1)^{-1} \sum_{m=0}^{\infty} m \chi(m, t); \quad (24)$$

$$\frac{\partial \chi(m, t)}{\partial t} = \sum_{j=-2}^2 D_j(m, m) e^{j\lambda\omega_1} \chi(m + j, t); \quad \chi(m, 0) = (m - \bar{n}) e^{-\omega_1 t m}; \quad (25)$$

$$\sum_{m=0}^{\infty} \chi(m, t) = 0.$$

In case of purely linear friction ( $\Gamma^{(2)} = 0$ ) a simple explicit expression may be obtained for  $N_1(t)$  by going from (25) to the moment equations  $\chi_k(t) = \sum_{m=0}^{\infty} m^k \chi(m, t)$  (the equation for  $\chi_k(t)$  does not contain subsequent moments). If the nonlinear friction is other than zero but small ( $\Gamma^{(2)} \ll \Gamma$ ) the iteration procedure results in an expression for  $N_1(t)$ :

$$N_1(t) = \bar{n}(\bar{n} + 1) e^{-2\Gamma t} - 16\bar{n}_2(\bar{n} + 1)^2 \Gamma^{(2)} t e^{-2\Gamma t}; \quad \Gamma^{(2)} \ll \Gamma; \quad \frac{1}{\Gamma^{(2)}} > t \gg \omega_1^{-1}. \quad (26)$$

As may be seen from (26) the correlator  $N_1(t)$  decreases non-exponentially in time (faster than  $\exp(-2\Gamma t)$ ) provided the nonlinear friction is taken into consideration.

The non-exponential dependence of  $N_1$  on  $t$  is particularly noticeable in case of purely nonlinear friction. Accordingly, the spectral representation  $N_1(\omega)$  is essentially non-Lorentzian. In the general case  $N_1(\omega)$  may be calculated numerically for any set of parameters  $\Gamma^{(2)}/\Gamma$  and  $\lambda\omega_1$  by solving (25) in the  $\omega$ -representation. In particular at high temperatures these equations are reduced to a differential equation

$$\left. \begin{aligned} & x(\Gamma + \gamma^{(2)}x) \frac{d^2 U}{dx^2} + [I(x + 1) + \gamma^{(2)}x(x + 2)] \frac{dU}{dx} + \\ & + \left[ \Gamma + 2\gamma^{(2)}x + i\frac{\omega}{2} \right] U = \frac{1}{2} (1 - x) \exp(-x); \\ & N_1(\omega) = \frac{1}{\pi(\lambda\omega_1)^2} \operatorname{Re} \int_0^{\infty} xU(x) dx; \quad \int_0^{\infty} U(x) dx = 0. \end{aligned} \right\} \quad (27)$$

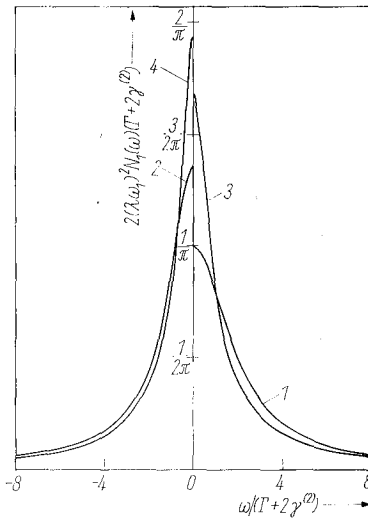


Fig. 5. Spectral distributions  $N_1(\omega)$  at high temperatures for different values of  $\gamma^{(2)}/\Gamma$ . The curves 1 to 4 refer to the following values of the parameter  $\gamma^{(2)}/\Gamma$ : 0; 2; 10; 100. The figure shows the right or left halves of the symmetrical curves

This equation being solved numerically<sup>1)</sup> it becomes possible to get the curves for  $N_1(\omega)$  for various ratios of  $\gamma^{(2)}$  to  $\Gamma$  (see Fig. 5). It is particularly evident from Fig. 5 that in the case of a purely nonlinear friction the peak is essentially narrower than the Lorentzian one.

## 6. Discussion of Results

It follows from (10) that in the considered model the nonlinear friction appears when the double frequency of the singled out oscillation lies in the range of frequencies of a continuous spectrum. This means that  $\Gamma^{(2)} = 0$  for high-frequency local oscillations but  $\Gamma^{(2)}$  may differ from zero for gap oscillations if  $\omega_{\kappa} < \omega_m/2$ . Nonlinear friction is especially important (as compared with a linear one) in case of weakly bound impurity centers when the amplitude of impurity oscillations prevails greatly that of the matrix atoms or when in the vicinity of  $2\omega_{\kappa}$  there is situated a peak in the density of states (that is a narrow band whose width is nevertheless larger than  $|\tilde{V}|, \Gamma$ ). The strong nonlinear friction arises also when the approximate equality  $2\omega_1 \approx \omega_2$  is fulfilled for two singled out oscillations. Then, if  $\omega_1 \gg \Gamma_2 \gg |V_{11}|, \Gamma_1, \Gamma_1^{(2)}$  and  $\Gamma_2 \gg |2\omega_1 - \omega_2|$ , the consideration given above is valid and  $\Gamma_1^{(2)} = V_{112}^2/\Gamma_2$  is particularly large (at  $\Gamma_2 \approx \approx |V_{11}| \approx |2\omega_1 - \omega_2|$  resonance effects appear which demand special investigation [10]). In similar conditions strong nonlinear friction of quasi-local oscillations also arises. If the interaction between singled out oscillations is essential ( $|2\omega_1 - \omega_2| \gg \Gamma_{1,2}$ ) then in accordance with (21) a significant nonlinear friction may appear at  $|\omega_1 \pm \omega_2| < \omega_m$  i.e. even in case of high-frequency local vibrations.

It may be concluded from the results obtained above that the nonlinear friction greatly influences the relaxation of singled out oscillations and changes their spectral distribution. Thus the dependence of widths of lines constituting the fine structure becomes quadratic in the number of the line  $m$  (instead of a linear dependence at a purely linear friction). At  $T \ll \omega_{\kappa}$  the nonlinear friction makes an exponentially small contribution to the main line ( $m = 1$ ) width but

<sup>1)</sup> The equation (27) was solved by the method of finite differences using the equality  $U(\infty) = 0$ . To satisfy the integral condition  $U(0)$  was determined by combining solutions corresponding to arbitrarily chosen  $U(0)$ . Such a scheme appears to be stable relative to this choice.

this contribution increases rapidly with rising temperature. In the classical limit the broadening due to nonlinear friction is proportional to temperature. In the case of quasi-local vibrations the linear friction is often almost independent of  $T$ . Therefore in this case the temperature increase leads to greater importance of both the nonlinear friction  $\gamma^{(2)} \sim T$  and the characteristic of non-equidistance  $v \sim T$ . At the same time the constant  $v/\gamma^{(2)}$  does not depend on  $T$  and the curves in Fig. 2 to 4 show actually the temperature dependence of corresponding parameters. The fact, obvious from Fig. 1 to 5, that the distribution becomes narrower in its central part with the growth of nonlinear friction is connected, in fact, to the decrease in the friction force with amplitude damping.

Nonlinear friction is due to terms of the type  $(a_n + a_n^+)^2 (a_k + a_k^+)$  in  $H_i$ . Such terms contribute also greatly to the integral intensity of the infrared absorption peak. It is difficult to investigate this problem with the method used here. It has been considered in a series of works for the non-resonance case and in [11] the possibility of resonance was taken into account.

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