Theory of nonlinear nonequilibrium oscillators interacting with a medium

M. I. Dykman

Institute of Semiconductors, Ukrainian Academy of Sciences

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The quantum-kinetic equation for the two-time correlation function of the operators of a nonequilibrium oscillator interacting wtih a medium is derived and solved for an arbitrary ratio between the vibrational-level nonequidistance due to nonlinearity of the oscillator to the level width. The polarizability and luminescence kinetics, total luminescence spectrum, and also the polarizability and luminescence under stationary conditions are considered. A criterion for amplification of light by an anharmonic oscillator under stationary conditions is found. The possibility of amplification by means of local and quasilocal oscillations in crystals is considered.

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The relaxation of a harmonic oscillator was investigated in a number of studies. This problem is of interest as an example of the establishment of equilibrium between a subsystem and a thermostat.[1-4] An examination of the dynamics of an oscillator interacting with a medium is encountered in some problems in the theory of lasers,[5-7] and also in the theory of local and quasilocal oscillations of impurities in crystals.[8] To explain the experimental results, however, it is frequently necessary to take into account the internal anharmonicity of the selected oscillator. For example, the radiation-oscillator anharmonicity due to the nonlinear terms in the polarizability of the medium governs the distinguishing features of the behavior of the propagation of light in nonlinear media.

The internal anharmonicity of local and quasilocal oscillations leads to asymmetry of the peaks in the spectra of infrared absorption and inelastic scattering of neutrons by these oscillations.[9] The asymmetry results from the fact that the vibrational levels of the nonlinear oscillator are not equidistant. If the deviation $\Delta\omega$ from equidistance is much larger than the reciprocal lifetime Γ, then the peak in the oscillator spectrum can have a fine structure whose individual lines correspond to transitions between different pairs of neighboring levels (the contributions from individual transitions cannot be identified in the spectrum of a harmonic oscillator). It should be noted that the non-equidistance becomes manifest in spectra of equilibrium oscillators only at finite temperatures, when the average occupation numbers of the excited levels differ from zero.

It is of interest to investigate the local and quasilocal oscillations under nonequilibrium conditions, for this makes it possible to study excited states of oscillations at low temperatures, when there is no thermal smearing. There is a special class of local oscillations that can exist only in a disequilibrium state, namely the strongly excited intramolecular oscillations in molecular crystals, which were considered by Ovchinnikov.[10] The localization of such oscillations is due to their internal anharmonicity, and their decay is made possible by the nonlinear interaction with phonons.

To determine the shapes of the peaks in the spectra of local and quasilocal oscillations it is necessary to calculate the spectral distribution of the correlation functions of the coordinates of these oscillations. To calculate the correlation functions if the equilibrium anharmonic oscillators interacting with the medium, Krivoglaz and the author[11] have obtained, on the basis of a previously-developed procedure,[12] a quantum-kinetic equation valid directly for the correlators. This procedure will be generalized in this paper to include the case of nonequilibrium anharmonic oscillators.

In Sec. 1 we obtain in explicit form the solution of the kinetic equation for the correlator of the operators of a nonequilibrium oscillator. This relation is used in Sec. 2 to find the time dependences of the polarizability and luminescence of the oscillators, and to investigate the complete luminescence spectrum. In Sec. 3 are analyzed the polarizability and luminescence of a nonequilibrium oscillator in the stationary regime. Criteria are obtained for amplification of light by an oscillator, and this amplification is analyzed. Section 4 contains concluding remarks. In particular, certain methods of oscillator pumping are considered.

1. CALCULATION OF CORRELATION FUNCTIONS

We consider a nonlinear oscillator of frequency ω interacting with a medium consisting of a set of harmonic oscillators. The oscillators of the medium are numbered by the subscript k and their frequencies ω_k form a quasicontinuous spectrum. We assume the interaction to be small enough, so that the relaxation time is $\Gamma^{-1} \gg \omega^{-1}$ and $\Gamma^{-1} \gg \omega_m^{-1}$, where ω_m is the maximum frequency of the continuous spectrum. This model describes well, for example, the frequency-nondegenerate local or quasilocal oscillations of a weakly bound impurity in a crystal.

The Hamiltonian of the system is

$$H=H_0+H_i, \quad H_0=\omega n+\frac{1}{2}Vn^2+\sum_k \omega_k n_k, \tag{1}$$

$$H_i=\sum_k V_k c c_k+\frac{1}{2}\sum_{kk'} H_{kk'}c_k c_{k'}; \quad H_{kk'}=V_{kk'}c+\tilde{V}_{kk'}c^2, \tag{2}$$

$$n=a^+a, \quad n_k=a_k{}^+a_k, \quad c=a+a^+, \quad c_k=a_k+a_k{}^+,$$

$\hbar = 1$. Here a, a^+ and a_k, a_k^+ are the annihilation and creation operators of the selected oscillations and of the continuous-spectrum oscillation. The parameters contained in H_i and the non-equidistance parameter V contain small constants in the assumed model.

We have left out of the Hamiltonian H_0 the cubic-anharmonicity term $\tilde{V}c^3$, which is rapidly oscillating and leads in the effects considered later on to the renormalization ($\sim \tilde{V}^2$) obtained in[13] for the parameters V and

ω. We shall assume this renormalization to have been effected (V = 0 if the oscillator is located in an inversion center). Nor does H_0 include small rapidly-oscillating terms such as Va^3a^+. The effect of the terms of the type $V_k c^2 c_k$, which have been omitted from formula (2), on the correlation functions of the equilibrium oscillators was considered in detail earlier.[11] If the density of the oscillations of the continuous spectrum in the vicinity of 2ω is equal to zero, then allowance for the terms $\tilde{V}_k c^2 c_k$ leads to addition of terms $\sim \tilde{V}_k^2$ to V and ω (the corresponding expressions are given in[12]). This renormalization will also be assumed effected in what follows.

Many properties of a nonequilibrium oscillator are determined by the two-time correlation function

$$\langle A(t+\tau)B(\tau)\rangle = \mathrm{Sp}[A(t)B\rho(\tau)], \tag{3}$$

where ρ is the density matrix of the system, and A and B are the operators of the selected oscillator. Assume that at the instant of time t = 0 the oscillator is described by a density matrix ρ_0 and the oscillations of the medium are in the state of thermodynamic equilibrium. To calculate the function $\langle A(t+\tau)B(\tau)\rangle$ at the times $t, \tau \gg \omega^{-1}, \omega_m^{-1}$ (it is precisely this time region which is usually of greatest interest, since it determines the spectral characteristics of the oscillator), we use a time-asymptotic perturbation theory, which is a generalization of the methods developed in[11,12] to include nonequilibrium problems.

We proceed to the interaction representation, introducing the operator $U(t) = e^{iH_0t}e^{-iHt}$. Calculating the trace in formula (3) for the complete system of eigenfunctions $(m|$ and $(m_k|$ of the harmonic oscillators in the occupation-number representation, accurate to terms quadratic in V_k, $V_{kk'}$, and $\tilde{V}_{kk'}$, we obtain

$$\langle A(t+\tau)B(\tau)\rangle = \sum^{\infty} A_{pq}B_{rs}(\rho_0)_{ln}\exp[i(E_p-E_q)t+i(E_m-E_s)\tau] \tag{4}$$
$$\times f(q,p;m,r;t)f(s,m;n,l;\tau),$$
$$E_m = \omega m + {}^1\!/_2 Vm^2,$$

where

$$f(q,p;m,r;t) = \prod_k (1+\bar{n}_k)^{-1} \sum_{\ldots,m_k,n_k,\ldots} (q,m_k|U(t)|r,n_k) \tag{5}$$
$$\times \exp\Big(-\nu\sum_k \omega_k n_k\Big)(m,n_k|U^{-1}(t)|p,m_k),$$
$$\nu = 1/k_BT,\ \bar{n}_k = [\exp(\nu\omega_k)-1]^{-1},$$

the sum in (4) is taken over all integer values of l, m, n, p, q, r, and s from zero to infinity. It is clear from (5) that the function f describes the oscillator relaxation due to the interaction with the medium. From (4) we can conclude that the individual oscillator operator matrix elements relax independently of the oscillator density-matrix elements. This result is essentially analogous to the results of Sec. 10 of the book of Faĭn and Khanin,[14] obtained from the condition of the diagonal singularity and the smallness of the interaction H_i under the assumption that certain effective initial conditions are satisfied. In the method proposed here there is no need for any special premises, and the calculation is carried out directly.

In the derivation of formula (4), expressions of the type

$$\sum_{\substack{\ldots,m_k,n_k,\ldots\\(m_k\neq n_k)}} \int (q,m_k|U(t)|r,n_k)(s,n_k|U(\tau)|l,m_k)$$
$$\times\exp\Big(-\nu\sum_k\omega_k m_k\Big)\exp\Big[i\tau\sum_k\omega_k(m_k-n_k)\Big]\Big\} \tag{6}$$

have been discarded, since they are proportional to integrals (with respect to ω_k) of terms that are rapidly-oscillating (owing to the last factor in (6)) at $\tau \gg \omega_m^{-1}$, and are of the order of Γ/ω, Γ/ω_m, $|d\Gamma/d\omega| \ll 1$ (this can be easily verified by expanding the operators U in (6) in powers of H_i). This approximation is valid if, as will be assumed from now on,

$$\mathrm{Sp}(AB\rho_0)\neq 0,\ \mathrm{Sp}[ABe^{-\nu\omega n}]\neq 0.$$

An equation for the function f can be obtained in the same manner as used in[11,12] for the analogous equilibrium function. Accurate to terms Γ/ω, $|V|/\omega$, $|d\Gamma/d\omega| \ll 1$ we have

$$\frac{\partial f(q,p;m,r;t)}{\partial t} = \sum_{j=-1}^{1} d_j(q,p)\exp[-iVj(q-p)t]f(q+j,p+j;m,r;t), \tag{7}$$
$$f(q,p;m,r;0) = \delta_{q,r}\,\delta_{p,m},$$

where

$$d_0(q,p) = -\Gamma(q)-\Gamma(p)-\gamma(q-p)^2-iP(q-p),$$
$$d_{\pm1}(q,p) = 2\Gamma(\bar{n}+{}^1\!/_2\pm{}^1\!/_2)[(q+{}^1\!/_2\pm{}^1\!/_2)(p+{}^1\!/_2\pm{}^1\!/_2)]^{1/2}, \tag{8}$$
$$\Gamma(q) = \Gamma[q(2\bar{n}+1)+\bar{n}],\ \bar{n} = [e^{\nu\omega}-1]^{-1}.$$

Here Γ and P are the real and imaginary parts of the polarization operator $R(\omega - i0)$,

$$R(\omega) = \sum_k \frac{2V_k^2\omega_k}{\omega^2-\omega_k^2} + \sum_{kk'} V_{kk'}^2\Big[\frac{(1+\bar{n}_k+\bar{n}_{k'})(\omega_k+\omega_{k'})}{\omega^2-(\omega_k+\omega_{k'})^2} + \frac{\bar{n}_{k'}-\bar{n}_k}{\omega-\omega_k+\omega_{k'}}\Big] \tag{9}$$
$$+\sum_k \tilde{V}_{kk}(2\bar{n}_k+1).$$

In this formula, account is taken, in first-order perturbation theory, of the terms of the fourfold anharmonicity in H_i. The main effect due to such phenomena in second-order perturbation theory is connected with the term proportional to γ in d_0, where

$$\gamma = 4\pi\sum_{kk'}\tilde{V}_{kk'}^2\bar{n}_k(\bar{n}_k+1)\delta(\omega_k-\omega_{k'}). \tag{10}$$

This term describes the modulation damping of the off-diagonal elements of f, which is connected with the elastic scattering of the continuous-spectrum oscillations by the selected oscillator; it is frequently decisive in the width of the spectral lines of high-frequency local oscillations.[9,15]

If the levels are equidistant (V = 0), then Eq. (7) coincides, accurate to terms with γ and P, with the density matrix considered in[4] for the damped harmonic oscillator. The presence of non-equidistance, however, greatly alters the manner in which equilibrium is established in the system. For example, if $|V| \gg \Gamma$, then the qualitative singularity of the oscillator relaxation, namely the connection of f(q, p; m, r; t) with f(q ± 1, p ± 1; m, r; t), practically drops out for the off-diagonal elements of f, since terms with j = 0 in (7) contain rapidly oscillating factors. The relaxation is then described by the usual expression for the energy-nondegenerate systems (the expression for $\Gamma(q)$ in (8) coincides with the half-width of the q-th level, calculated in accordance with the usual Weisskopf and Wigner perturbation theory).

In the general case of an arbitrary ratio $|V|/\Gamma$, Eq. (7) is conveniently solved by the method of generating functions.[12,4] For the function

$$f_k(x,t;m,r)=\sum_{\beta=0}^{\infty}\left(\frac{\alpha!}{\beta!}\right)^{1/2}f(q,p;m,r;t)x^{\beta}\exp\left[iPkt+\frac{1}{2}iV(p^2-q^2)t\right];$$

$$\alpha=\max(p,q);\quad \beta=\min(p,q);\quad k=q-p, \tag{11}$$

we can obtain from (7) a linear partial differential equation of first order, the solution of which is of the form

$$f_k(x\ t;m,r)=\left[\operatorname{ch}a_kt+\frac{2\Gamma(2\bar{n}+1)-4x\Gamma\bar{n}+iVk}{2a_k}\operatorname{sh}a_kt\right]^{-|k|-1} \tag{12}$$

$$\times\exp\left[-\left(\gamma k^2-\frac{1}{2}iVk-\Gamma\right)t\right]f_{k0}(x,t;m,r),$$

$$a_k^2=\Gamma^2+i\Gamma Vk(2\bar{n}+1)-{}^1\!/_4V^2k^2,$$

where

$$f_{k0}(x,t;m,r)=\left\{\frac{x[a_{k1}-a_{k2}\exp(2a_kt)]-a_{k1}a_{k2}[1-\exp(2a_kt)]}{x[1-\exp(2a_kt)]-[a_{k2}-a_{k1}\exp(2a_kt)]}\right\}^{\eta} \tag{13}$$

$$\times\left(\frac{\xi!}{\eta!}\right)^{1/2}\delta_{k,r-m};\quad \xi=\max(m,r);\quad \eta=\min(m,r);$$

$$a_{k1,2}=\frac{2\Gamma(2\bar{n}+1)+iVk\pm 2a_k}{4\Gamma\bar{n}}.$$

Expressions (4) and (11)—(13) enable us to find the two-time correlation functions of the nonequilibrium oscillator interacting with a medium at an arbitrary ratio of the non-equidistance of the vibrational levels to the damping, and at arbitrary temperatures.

2. INTERACTION OF NONLINEAR NONEQUILIBRIUM OSCILLATORS WITH RADIATION

We consider the linear reaction of a nonequilibrium oscillator to an electric field. Greatest interest attaches to the resonance region, when the field frequency Ω is close to the oscillator frequency ω. For a non-equilibrium oscillation, which is described at the instant t_0 by a density matrix ρ_0, we can introduce a polarizability $\chi(\Omega, t-t_0)$ that depends on the time and on the frequency, such that

$$\langle M(t)\rangle_E=\mu^2\chi(\Omega,\ t-t_0)E_\Omega e^{-i\Omega t},$$

where $\langle M(t)\rangle_E$ is the field-dependent part of the oscillator dipole moment (a nonequilibrium oscillator can have a dipole moment also in the absence of a field), E_Ω is the intensity of the field at the frequency Ω, and μ is a coefficient that determines the dipole moment of the oscillator $M = \mu(a + a^+)$. It is easy to show that

$$\chi(\Omega,t-t_0)=i\int_{t_0}^{t}e^{i\Omega(t-\tau)}\chi(t-\tau,\tau-t_0)d\tau,\quad \chi(t,\tau)=\operatorname{Sp}\{[c(t),c]\rho(\tau)\}. \tag{14}$$

In the calculation of lasers in plasma theory and for a number of other applications, it is frequently necessary to know the power $w_\Omega(t-t_0)$ drawn by the field form the non-equilibrium oscillator excited at an instant t_0.[1)] If the oscillator is in a classical field of intensity $E = E_\Omega e^{-i\Omega t} + E_\Omega^* e^{i\Omega t}$, then, accurate to the rapidly-oscillating terms,

$$w_\Omega(t-t_0)=-2\Omega\mu^2|E_\Omega|^2\operatorname{Im}\chi(\Omega,\ t-t_0). \tag{15}$$

Formula (15) allows us to investigate the spectrum of the absorption (amplification) of the light by the non-equilibrium oscillator.

To investigate the luminescence spectrum it is necessary to consider the interaction of the oscillator with equilibrium radiation having the temperature of the medium. In this case the power radiated by the non-equilibrium oscillation is determined by the expression

$$W(t)=\int_0^{\infty}\eta(\Omega)d\Omega\,\mu^2\Omega Q(\Omega,t-t_0), \tag{16}$$

$$Q(\Omega,t-t_0)=2\operatorname{Re}\int_{t_0}^{t}d\tau\,e^{i\Omega(t-\tau)}q_\Omega(t-\tau,\tau-t_0)$$

$$q_\Omega(t,\ \tau)=\operatorname{Sp}\{\rho(\tau)[(\bar{n}_\Omega+1)cc(t)+\bar{n}_\Omega c(t)c]\}.$$

Here $\bar{n}_\Omega$ is the Planck number. The function $\eta(\Omega)$ is determined by the photon state density, so that

$$\langle E^2\rangle=\int_0^{\infty}\eta(\Omega)d\Omega(2\bar{n}_\Omega+1).$$

It is assumed henceforth that $\eta(\Omega)$ is a smooth function in the vicinity of $\Omega \sim \omega$.

It is seen from (14)—(16) that an analysis of the interaction of a non-equilibrium oscillator with radiation entails the calculation of two-time correlation functions, which was carried out in general form above.

Since we are considering the resonance region $\Omega \sim \omega$, it is necessary to retain in $\chi(t, \tau)$ and $q_\Omega(t, \tau)$ in the calculation of the shapes of the absorption and luminescence peaks, only the terms that comprise products of $e^{-i\omega t}$ by a smooth function of t and τ. To this end we take into account the term a(t) in the operator c(t), the term a^+ in c, and only the diagonal elements in $\rho(\tau)$. This approximation is valid if

$$\Gamma|\langle a^2\rangle_0|<\omega|\langle aa^+\rangle_0|,\ \langle A\rangle_0=\operatorname{Sp}(\rho_0A).$$

Using formulas (4) and (11)—(13) as well as the analogous expressions for the function $\langle B(\tau)A(t + \tau)\rangle$ and taking into account the explicit form of the operators, we obtain accurate to terms Γ/ω, $|V|/\omega$, and $|d\Gamma/d\omega|$

$$\chi(t,\tau)=\exp\left[-i\tilde{\omega}t-\left(\gamma-i\frac{V}{2}-\Gamma\right)t\right]\psi^{-2}(t)\sum_{m=0}^{\infty}\rho_m$$

$$\times[1-(\bar{n}+m+1)\varphi(t,\tau)][1-(\bar{n}+1)\varphi(t,\tau)]^{m-1}[1-\bar{n}\varphi(t,\tau)]^{-m-2},$$

$$\varphi(t,\tau)=i\frac{V}{a}\psi^{-1}(t)\operatorname{sh}ate^{-2\Gamma\tau},\quad \psi(t)=\operatorname{ch}at+\left[1+i\frac{V}{2\Gamma}(2\bar{n}+1)\right]\frac{\Gamma}{a}\operatorname{sh}at,$$

$$a^2\equiv a_1^2=\Gamma^2+i\Gamma V(2\bar{n}+1)-V^2/4,\quad \tilde{\omega}=\omega+P,\quad \rho_m=(m|\rho_0|m). \tag{17}$$

Without going into details of the analysis of this formula, we note that the polarizability of the harmonic oscillator does not depend on its initial state, inasmuch as at V = 0 we have

$$\varphi(t,\tau)=0,\quad \sum_{m=0}^{\infty}\rho_m=1,\quad \chi(t,\tau)=\exp[-i\tilde{\omega}t-(\gamma+\Gamma)t].$$

As $\tau \to \infty$, the quantity $\chi(t, \tau)$ tends to the equilibrium polarizability obtained in[12] for a nonlinear oscillator. However, whereas the nonequidistance of the levels manifests itself in the polarizability of the equilibrium operator only at finite temperatures, $\bar{n} > 0$,[12] the polarizability of a nonequilibrium oscillator, as seen from (17) is affected by the nonequidistance also at $\bar{n} = 0$. This is understandable, since the nonequidistance manifests itself when there are transitions between several oscillator levels.

In the resonance region, assuming $\bar{n}_\Omega = \bar{n}$, we obtain for the function $q_\Omega(t, \tau)$

$$q_\Omega(t,\tau)=e^{-2\Gamma\tau}\exp[-i\tilde{\omega}t-(\gamma-{}^1/{}_2iV-\Gamma)t]\psi^{-2}(t)\sum_{m=0}^{\infty}\rho_m[1-(\bar{n}+1)\varphi(t,\tau)]^{m-1}$$
$$\times[1-\bar{n}\varphi(t,\tau)]^{-m-2}[m-\bar{n}+\bar{n}(\bar{n}+1)\varphi(t,\tau)]. \quad (18)$$

Formulas (16) and (18) describe the time damping of the luminescence of a nonlinear nonequilibrium oscillator.

Let us see how the energy radiated by the oscillator is distributed over the frequency during the entire relaxation time, i.e., let us calculate the shape of the peak in the luminescence spectrum of the oscillator. As is clear from (16), the shape of the peak is determined, accurate to the smooth factor $\eta(\Omega)\Omega$, by the function

$$Q(\Omega)=\int_{t_0}^{\infty}Q(\Omega,t-t_0)\,dt=2\,\mathrm{Re}\int_0^{\infty}dt\int_0^{\infty}d\tau\,e^{i\Omega t}q_\Omega(t,\tau). \quad (19)$$

From (19) with allowance for (17) and (18) we obtain

$$Q(\Omega)=\frac{1}{\Gamma}\,\mathrm{Im}\int_0^{\infty}dt\exp\left[i\left(\Omega-\tilde{\omega}+\frac{V}{2}\right)t-(\gamma-\Gamma)t\right]\psi^{-1}(t)$$
$$\times\frac{a}{V\,\mathrm{sh}\,at}\sum_{m=0}^{\infty}\rho_m\left\{1-\frac{[1-(\bar{n}+1)\varphi(t,0)]^m}{[1-\bar{n}\varphi(t,0)]^{m+1}}\right\}. \quad (20)$$

This formula gives the complete solution of the problem of the luminescence spectrum of a nonequilibrium oscillator at an arbitrary ratio of the nonequidistance of the levels $|V|$ and their width and for arbitrary temperatures. The integrated intensity of the peak, accurate to Γ/ω, is equal to $\Gamma^{-1}\pi\langle n\rangle_0-\bar{n})$, and is independent of V or γ. These parameters, however, do determine the complicated asymmetric shape of the peak, which can be obtained by numerical integration with the aid of formula (2) at arbitrary values of the parameters.

At V = 0, the function $Q(\Omega)$ has a Lorentz shape with half-width $\Gamma+\gamma$ and with a maximum at $\Omega=\tilde{\omega}$. With increasing ratio $|V|/\Gamma$, the line broadens, the position of the maximum shifts, the asymmetry increases, and at $|V|\gg\Gamma(2\bar{n}+1)$ a fine structure appears in the spectrum.

At $|V|\gg\Gamma(2\bar{n}+1)$, accurate to terms $\sim\Gamma/|V|$, we have

$$Q(\Omega)=\frac{1}{\Gamma}\sum_{s=0}^{\infty}\frac{\Gamma_s}{\Gamma_s^2+\Omega_s^2}\left\{\sum_{m=s+1}^{\infty}\rho_m-e^{-\omega\nu(s+1)}\right\}+\frac{2}{V}\sum_{s=0}^{\infty}\frac{\Omega_s}{\Gamma_s^2+\Omega_s^2}$$
$$\times\left\{(2\bar{n}+1)\left[\sum_{m=s+1}^{\infty}\rho_m-e^{-\omega\nu(s+1)}\right]+(s+1)[2e^{-\omega\nu(s+1)}-\bar{n}\rho_s-(\bar{n}+1)\rho_{s+1}]\right\},$$
$$\Omega_s=\Omega-\tilde{\omega}-V(s+{}^1/{}_2),\quad \Gamma_s=2\Gamma(s+1)(2\bar{n}+1)-\Gamma+\gamma. \quad (21)$$

The first term in this expression describes the aggregate of Lorentz lines. The half-width Γ_s of the s-th line is equal (apart from the modulation broadening) to the sum of the half-widths of the levels s and s + 1 (see (8)), and its position is determined by the frequency of the transition between these levels. The amplitude of the s-th line is proportional to the difference between the sum of the populations of all the low-lying levels and its equilibrium value (this is due to the fact that the transitions occur only between neighboring levels of the relaxing oscillator).

The second term in (21) is proportional to $\Gamma/|V|$. It describes the corrections to the calculation of the luminescence spectrum in accord with the usual theory of Weisskopf and Wigner, which are connected with the partial degeneracy of the oscillator spectrum with respect to frequency. When the corrections are taken into account, the fine-structure line shape becomes asymmetrical, and the line intensities depend not only on the populations of the higher-lying levels, but also on the population of the level to which the transition takes place.

Since the line widths increase with temperature and the fine structure becomes smeared out, it is of interest to consider the luminescence spectrum of the oscillator at relatively low temperatures, when $\bar{n}=0$. Then

$$Q(\Omega)=\Gamma^{-1}\,\mathrm{Re}\sum_{s=0}^{\infty}(\Gamma_s-i\Omega_s)^{-1}\sum_{m=s+1}^{\infty}\rho_m\sum_{n=s+1}^{m}\binom{m}{n}\left(\frac{2\Gamma}{2\Gamma+iV}\right)^{m-n}\left(\frac{iV}{2\Gamma+iV}\right)^{n-1} \quad (22)$$

(the values of all the parameters are taken here at $\bar{n}=0$). It is clear from this formula that at finite V/Γ the concept of the line corresponding to an individual transition between the levels becomes arbitrary. The amplitude of the line is complex and it is determined by the interference of the occupation numbers of all the upper (and at $\bar{n}\neq0$ also the lower) levels. In particular, in the spectrum of the harmonic oscillator there is effectively excited at $\bar{n}=0$ only one line with s = 0.

3. INTERACTION OF OSCILLATORS WITH RADIATION IN THE STEADY STATE

The investigation of the steady states of non-equilibrium systems is of interest for a number of applications, especially in laser physics. The explicit expressions (14)—(18) for the absorption and luminescence spectra of non-equilibrium nonlinear oscillators make it possible to analyze completely any regime, including the steady state. To obtain the observable quantities $\chi(\Omega,t-t_0)$ and $W(t)$ it is necessary to average in (14)—(16) with respect to t_0 with a certain weighting function $\zeta(t_0)$ having the meaning of the probability density for the appearance of an oscillator with a density matrix ρ_0 at an instant t_0. The function $\zeta(t_0)$, just as ρ_0, is determined by the external system that excites the oscillator.

In the steady state, the only singled-out instant of time is the observation instant t, and consequently $\zeta(t_0)$ depends on the difference $t-t_0$. We note that in the stationary regime the function $\zeta(t-t_0)$ is equal to the probability density of an event in which the oscillator appears for the last time prior to the observation at an instant separated by $t-t_0$ from the observation instant. Using the normalization condition

$$\int_{-\infty}^{t}\zeta(t-t_0)\,dt_0=1 \quad (23)$$

and Eqs. (14) and (16)—(18), we can show that, accurate to Γ/ω, the integral intensity of the absorption peak in the stationary regime is

$$\int_0^{\infty}d\Omega\,\mathrm{Im}\,\chi_{\mathrm{st}}(\Omega)=\pi,$$

and the integral intensity of stationary-luminescence peak is

$$\int_0^{\infty}d\Omega\,Q_{\mathrm{st}}(\Omega)=2\pi(\langle n\rangle_0-\bar{n})\int_0^{\infty}\zeta(t)e^{-2\Gamma t}\,dt.$$

From these expressions, if (15) is taken into account, we see that, relative to the entire spectrum, the oscillator in any steady state absorbs the energy of the applied field independently of the pump, of the non-equidistance of the levels, and of the temperature. It will be shown later on, however, that under certain conditions, there is an interval of the frequencies Ω where $\operatorname{Im}\chi_{st}(\Omega) < 0$, i.e., amplification of the field takes place. The intensity of the luminescence peak is likewise independent of the anharmonicity, but the pump and the temperature, as expected, do affect this intensity.

If the excitation process constitute shot noise with an average frequency λ (e.g., scattering of some particles by the oscillator), then $\xi(t-t_0) = \lambda\exp[-\lambda(t-t_0)]$. Let us consider in this case the imaginary part of the oscillator polarizability,

$$\chi_\lambda(\Omega)=\lambda\operatorname{Re}\int_0^\infty dt\int_0^\infty d\tau\, e^{i\Omega t-\lambda(t+\tau)}\chi(t,\tau). \tag{24}$$

Expression (17) allows us to obtain $\chi_\lambda(\Omega)$ for all temperatures, at arbitrary V/Γ, λ/Γ, and ρ_0, and thus calculate completely the shape of the peak of light absorption by a nonlinear oscillator in the steady state. The actual calculation is easily carried out with a computer. It is possible to treat analytically several limiting cases.

At small nonequidistance, when $|V| \ll \Gamma$, we have in first order in V/Γ

$$\chi_\lambda(\Omega)=\frac{\Gamma+\gamma+\lambda}{(\Gamma+\gamma+\lambda)^2+\Delta^2}+\frac{\lambda V}{\Gamma(2\Gamma+\lambda)}\left[\frac{\Delta}{(\Gamma+\gamma+\lambda)^2+\Delta^2}-\frac{\Delta}{(3\Gamma+\gamma+\lambda)^2+\Delta^2}\right]\times(\langle n\rangle_0-\bar{n}), \tag{25}$$

$\Delta = \Omega - \tilde{\omega} - \frac{1}{2}V(4\bar{n} + 1)$. The peak of the spectral distribution of a nonequilibrium oscillator is asymmetrical, as seen from (25), and the asymmetry comes into play in first order in V/Γ (for an equilibrium oscillator, it is proportional to V^3/Γ^3[12]) and increases with the pump. At $|V| \ll \Gamma$ the oscillator absorbs the energy of the applied field at all frequencies. Neglecting the nonequidistance, the distribution of $\chi_\lambda(\Omega)$ turns out to be of the Lorentz type, and depends only on one pump parameter, namely the average excitation frequency λ, which enters as an increment to the absorption-line half-width. The quantity λ is added also to the half-width of the Lorentz line of the stationary luminescence of the harmonic oscillator. At $V \neq 0$, the luminescence spectrum becomes asymmetrical, and a fine structure appears in it at $|V| \gg \Gamma$.

Expressions (24) and (17) for $\chi_\lambda(\Omega)$ become much simpler in the case of low temperatures. At $\bar{n} = 0$ we have

$$\chi_\lambda(\Omega)=\lambda\operatorname{Re}\sum_{s=0}^{\infty}(\Gamma_s+\lambda-i\Omega_s)^{-1}\sum_{m=s}^{\infty}\binom{m}{s}\rho_m\sum_{n=0}^{m-s}\binom{m-s}{n}\frac{(-1)^n(n+s+1)}{\lambda+2\Gamma(n+s)}\times\left(\frac{iV}{2\Gamma+iV}\right)^{n+s}, \tag{26}$$

where the Γ_s were calculated with allowance for the condition $\bar{n} = 0$.

Formula (26) demonstrates the interference of the transitions between the levels of the nonlinear oscillator. If $|V| \gg \Gamma$, then the individual transitions occur almost independently, and we can obtain in first order in Γ/V

$$\chi_\lambda(\Omega)=\frac{\lambda}{2\Gamma}\left\{\sum_{s=0}^{\infty}\frac{\Gamma_s+\lambda}{(\Gamma_s+\lambda)^2+\Omega_s^2}\sum_{m=s}^{\infty}\rho_m[\delta_{m,s}+C(\lambda;m,s)]-\frac{2\Gamma}{V}\sum_{s=0}^{\infty}\frac{\Omega_s}{(\Gamma_s+\lambda)^2+\Omega_s^2}\sum_{m=s}^{\infty}\rho_m\left[m(\delta_{m,s}-\delta_{m,s+1})-\frac{\lambda}{2\Gamma}C(\lambda;m,s)\right]\right\},$$

$$C(\lambda;m,s)=\left(1-\frac{\lambda}{2\Gamma}\right)\frac{m!}{s!}\prod_{j=0}^{m-s}\left(\frac{\lambda+2\Gamma_s}{2\Gamma}+j\right)^{-1}. \tag{27}$$

This leads to a criterion for the amplification of light by a nonlinear non-equilibrium oscillator in the case of stationary excitation of the shot-noise type

$$\lambda>2\Gamma\quad(|V|\gg\Gamma). \tag{28}$$

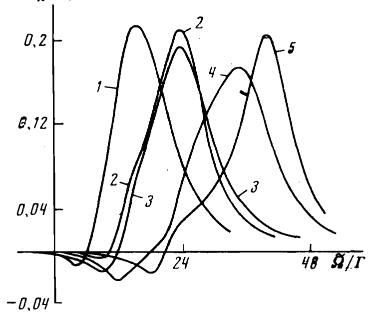
If (28) is satisfied we have $C(\lambda; m, s) < 0$ and the principal term in $\chi_\lambda(\Omega)$ can be negative. It is interesting that the amplification criterion is independent, accurate to terms $\sim\Gamma/V$, of the pump power, i.e., of the energy that the oscillator acquires during one excitation act, and indicates only that the average excitation frequency should exceed the characteristic reciprocal relaxation time 2Γ. The amplitudes of the fine-structure lines that are asymmetrical when the second term of (27) is taken into account, and in particular of the lines corresponding to amplification, depends on the pump power, which is characterized by the matrix ρ_0, and the amplification increases with increasing pump.[2)]

The qualitative picture of the change of $\chi_\lambda(\Omega)$ with changing λ, ρ_m, and V/Γ at $\bar{n} = 0$ is shown in the figure, where it is assumed for simplicity that $\gamma = 0$ (this corresponds, for example, to the case of very low temperatures $T \ll \omega_m$). When comparing curves 1 with 4, and 2 with 5, on which λ and ρ_m are the same, we see that with increasing nonequidistance the gain and the amplification frequency region increase; they increase also with increasing pump frequency λ (curves 2 and 3), and especially with increasing pump power (curves 2 and 4) (it has turned out, for example that if $V/\Gamma = 6$ and the pump goes to the first level, $\rho_1 = 1$, then no gain is produced at any value of λ).

In the derivation of (24)—(28) we have assumed that as a result of each excitation act the diagonal elements of the density matrix of the oscillator ρ_0 turn out to be the same, i.e., in fact averaging was carried out over the state of the oscillator immediately after the excitation, independently of the averaging with respect to the instant of excitation. It is clear that this is valid if the energy spectrum of the excitation is broad enough ($\gtrsim \omega$). However, even under more stringent assumptions these averagings are relatively independent at $\lambda \ll \omega$, if the duration of the excitation act is much shorter than λ^{-1} or Γ^{-1}, since the final state is determined by the phase of the initial one (by the rapidly-oscillating off-diagonal elements of the oscillator density matrix).

4. DISCUSSION OF THE RESULTS

The theory of oscillator relaxation and the time-dependent linear theory of the interaction of a non-equilibrium nonlinear oscillator with light, which were



Dependence of the imaginary part of the polarizability of the nonlinear oscillator in the stationary regime on the frequency $\tilde{\Omega} = \Omega - \tilde{\omega} - V/2$ at different pumps and different ratios V/Γ ($\bar{n} = 0$). The curves correspond to the following values of the parameters (in a scale in which $\Gamma = 1$): 1—$\lambda = 2.5$, $V = 6$, $\rho_2 = 1$; 2—$\lambda = 2.5$, $V = 12$, $\rho_1 = 1$; 3—$\lambda = 4$, $V = 12$, $\rho_1 = 1$; 4—$\lambda = 2.5$, $V = 12$, $\rho_2 = 1$; 5—$\lambda = 2.5$; $V = 20$, $\rho_1 = 1$.

developed in this paper, can be applied to a large number of physical systems. The relaxation theory describes, e.g., laser radiation in nonlinear media, particularly in a plasma. An example of nonlinear oscillators can be electrons and holes in a quantizing transverse magnetic field in thin films or in narrow near-surface channels of a semiconductor, when a strong electric field leads to quantization of the motion transverse to the surface and the corresponding band splitting is large in comparison with the cyclotron frequency. A nonequilibrium state in the system can be obtained by abruptly turning on an electric or a magnetic field. The anharmonicity of the oscillators is due to the nonparabolic dispersion of the carriers, and the energy relaxation is due to several mechanisms, included among which is the inelastic scattering of the carriers by the phonons. If this mechanism predominates, then certain results of this paper (in particular, the individual results concerning the amplification of the light) can be applied also to such systems if there is no degeneracy in them (we did not consider the translational motion of the carriers along the surface).

All the results obtained in this paper are directly applicable to local and quasilocal oscillations of weakly bound impurities in crystals, and to nonequilibrium local oscillations in pure molecular crystals (see [10]). The characteristic frequencies ω of the local and quasilocal oscillations of the impurities amount to 10^{13}—10^{14} $\sec^{-1}$, and the widths Γ at low temperatures can be 10^{10}—10^{11} $\sec^{-1}$ (according to estimates by Ovchinnikov, [10] for a strongly-excited local oscillation of a molecular crystal the value of Γ can be 10—10^{5} $\sec^{-1}$). An investigation of the transient processes in systems of local and quasilocal oscillations can be carried out with the aid of two short (of duration $< \Gamma^{-1}$) pulses, by using the first (high-power) pulse as a pump and measuring the absorption or scattering of the second (weak) pulse as a function of the interval between them. Experimental installations of this kind with picosecond pulses are already in existence (see, e.g., [16]), and a theoretical analysis of the results on the basis of formulas (14), (15) and (17) makes it possible not only to determine all the parameters of the investigated oscillators, but also to find directly the change of the state of the nonlinear oscillator in the field of a strong wave (the matrix ρ_0).

The density matrix ρ_0 can be determined also from the luminescence of the selected oscillations. Since either local or quasilocal oscillations are frequently excited in the impurity center in the case of an electronic transition, [17] it follows that the determination of ρ_0 from their emission will make it possible to reconstruct the essential details of the electron-phonon interaction.

The development of steady-state lasers operating on local and quasilocal oscillations calls for a high pump frequency $\lambda \gtrsim 2\Gamma$ (formula (28)). The stationary excitation by a neutron or electron flux with sufficiently large λ is a complicated task, since large particle flux densities are needed. A feasible way of exciting quasilocal or local oscillations is by optical means. For real impurity local or quasilocal oscillations, values $\lambda \sim 10^{10}$ $\sec^{-1}$ can be obtained at a stationary pump field 10^4—10^5 V/cm (it is desirable that the laser pump frequency coincide with one of the secondary peaks in the impurity-oscillation spectrum). The stationary pumping can also be effected with the aid of periodic high-power picosecond pulses. The polarizability and luminescence in such a regime are determined by expressions (14)—(18) and by the excitation-probability function $\zeta(t - t_0) = T^{-1}\theta(T - t + t_0)$, where T is the pulse repetition period.

In the case of wide-band semiconductors at very low temperatures, pumping with high λ can be obtained if the local oscillations are connected with the impurities that with which the strong-field-activated [18] hopping conductivity is realized.

We note that inhomogeneous broadening of the spectra of the local oscillations in impurity crystals is of no significance from the point of view of amplification if the frequency region of the amplification exceeds the inhomogeneous broadening. To increase the amplification region it is necessary, as seen from the figure, to increase the average energy of the oscillator.

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[1] Expressions (14) and (15) for $\chi(\Omega, t-t_0)$ and $w_\Omega(t-t_0)$ are valid also in the case when a wave of frequency Ω is incident on the nonequilibrium oscillator at the instant t_0. In this case ρ_0 depends on t_0-t_1, where t_1 is the instant of excitation of the oscillator.

[2] The terms $\sim\Gamma/V$ were omitted in the derivation of the amplification criterion (28). When $\langle n\rangle_0 \gg 1$ (large pump power, these terms can be significant and the condition (28) becomes less stringent. We note also that other pumping regimes correspond to other amplification criteria. In particular, if $\zeta(t-t_0) = \delta(T - t + t_0)$, then even in the harmonic-oscillator spectrum there are frequency intervals corresponding to amplification.

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