ment from the ESR data obtained under axial pressure and is clearly the only method for the determination of this quantity.

[1]N. G. Kabanova, S. N. Lukin, G. N. Neilo, and L. F. Chernysh, Kristallografiya 21, 1235 (1976) [Sov. Phys. Crystallogr. 21, 716 (1976)].
[2]M. M. Zaitov, Prib. Tekh. Eksp. No. 4, 194 (1969).
[3]A. A. Galkin, S. N. Lukin, and G. A. Tsintsadze, Phys. Status Solidi B 71, 205 (1975).
[4]R. W. G. Wyckoff, Crystal Structures, Vol. 3, Interscience, New York (1951).
[5]A. A. Galkin, A. Yu. Kozhukhar', and G. A. Tsintsadze, Zh. Eksp. Teor. Fiz. 70, 248 (1976) [Sov. Phys. JETP 43, 128 (1976)].
[6]V. N. Vasyukov, S. N. Lukin, and G. A. Tsintsadze, Fiz. Nizk. Temp. 3, 1476 (1977) [Sov. J. Low Temp. Phys. 3, 710 (1976)].
[7]E. Feher and M. D. Sturge, Phys. Rev. 172, 244 (1968).
[8]A. G. Danilov, J. C. Vial, and A. Manoogian, Phys. Rev. B 8, 3124 (1973).
[9]G. Simmons and H. Wang, Single Crystal Elastic Constants and Calculated Aggregate Properties (Handbook), MIT Press, Cambridge, Mass (1971), p. 3.
[10]E. M. Voronkova, B. N. Grechushnikov, G. I. Distler, and I. P. Petrov, Optical Materials for Infrared Technology [in Russian], Nauka, Moscow (1965).
[11]R. B. Hemphill, P. L. Donoho, and E. D. McDonald, Phys. Rev. 146, 329 (1966).
[12]S. A. Al'tshuler and B. M. Kozyrev, Electron Paramagnetic Resonance of Compounds of Intermediate-Group Elements [in Russian], Nauka, Moscow (1972), p. 274.
[13]A. M. Germanier and R. Lacroix, Helv. Phys. Acta 34, 401 (1961).

Translated by A. Tybulewicz

Heating and cooling of local and quasilocal vibrations by a nonresonance field

M. I. Dykman

Institute of Semiconductors, Academy of Sciences of the Ukrainian SSR, Kiev

(Submitted January 25, 1978)

Fiz. Tverd. Tela (Leningrad) 20, 2264–2272 (August 1978)

The dynamics of an anharmonic oscillator interacting with phonons in a nonresonant quasimonochromatic field is studied. The field-induced decay effects alter considerably the effective oscillator temperature. When an induced decay corresponds to simultaneous excitation of the oscillator and creation of a phonon, a runaway effect occurs for strong pumping, i.e., the oscillator energy increases exponentially with time. Mechanisms limiting the runaway effect are proposed. The limiting mechanisms lead to an inverse population of the oscillator levels in a certain energy interval. The luminescence and absorption spectra corresponding to local and quasilocal vibrations are studied. For an inverted population, the anharmonic vibrations can amplify light near the fundamental absorption frequency. If the frequency of the pumping field is higher than the oscillator frequency, the pumping field amplifes light and phonons at the difference frequency.

PACS numbers: 63.10. + a

The study of the dynamics of localized vibrations of impurities in crystals (local or quasilocal vibrations) is dependent on the solution of a more general problem of an oscillator interacting with a medium.[1] The relaxation of a harmonic oscillator whose interaction with a medium can be described by an effective friction force $\Gamma q_\varkappa$ [$q_\varkappa = \sqrt{\hbar/2\omega_\varkappa}(a_\varkappa + a_\varkappa^+)$ is the normal coordinate of an oscillator $\varkappa$ and $\omega_\varkappa$ is its frequency] and by a random force was studied in Refs. 2–5. A quantum-mechanical transport equation corresponding to such a model was discussed in Ref. 4. It was shown in Ref. 4 that, after a time $t \gg \Gamma^{-1}$ from the application of a regular external force when the initial distribution $\rho(0)$ is "forgotten," the density matrix $\rho(t)$ represents a shifted Boltzmann distribution. The shifted distribution can be obtained from the equilibrium distribution by the following simple unitary transformation:

$$\rho(t) = D(t)\,\rho_{eq} D^+(t);\ \rho_{eq} = [\bar{n}(\omega_\varkappa) + 1]^{-1} \exp(-\lambda\omega_\varkappa a_\varkappa^+ a_\varkappa), \quad (1)$$
$$\lambda = \frac{1}{T},\ \bar{n}(\omega) = [e^{\lambda\omega} - 1]^{-1},$$
$$D(t) = \exp[v(t)\,a_\varkappa^+ - v^*(t)\,a_\varkappa];$$

$$v(t) = i\int_0^t dt' f(t') \exp[-(\Gamma + i\omega_\varkappa)(t - t')],\ \hbar = 1,$$

where f(t) is an external force (measured in frequency units).

In fact, Eq. (1) holds only near a resonance when $|\omega - \omega_\varkappa| \ll \omega_m$, where ω is the characteristic frequency of the applied force, ω_m is the characteristic frequency of variation of the polarization operator of the oscillator $R(\omega)$, and $\Gamma = \mathrm{Im}\,R(\omega_\varkappa)$. For $|\omega - \omega_\varkappa| \sim \omega_m$ the dispersion of $R(\omega)$ influences strongly the oscillator dynamics in an external field. For example, it follows from Eq. (1) that the weak-field absorption cross section $\sigma(\omega) \propto \Gamma/(\omega - \omega_\varkappa)^2 (|\omega - \omega_\varkappa| \gg \Gamma)$ decreases monotonically as a function of the frequency detuning, whereas the correct expression $\sigma(\omega) \propto \mathrm{Im}\,R(\omega)/(\omega - \omega_\varkappa)^2$ (see, for example, Ref. 6) has an additional structure near $|\omega - \omega_\varkappa| \sim \omega_m$. Such a structure manifests itself in the infrared absorption spectra of local vibrations[7]; in addition to a central peak at $\omega = \omega_\varkappa$, the absorption cross section $\sigma(\omega)$ exhibits side bands that are shifted relative to $\omega_\varkappa$ through distances approximately corresponding to the

0038-5654/78/08 1306-06 $02.40

maxima in the phonon density of states. For a strong field and $\omega \sim |\pm\omega_x \pm \omega_m|$, the decay effects involving the simultaneous creation or annihilation of a photon, a phonon, and a quantum of local vibrations can dominate the relaxation of local vibrations.

The distribution of vibrations over excited states in strong fields depends strongly on whether the field frequency is higher than the oscillator frequency and the frequency of phonons participating in the decay or not. In the latter case, the decay can occur only when either the oscillator or phonons are excited. The oscillator is then coupled to a distribution of phonons participating in the relaxation. This can be described by an effective temperature $T^* = T\frac{\omega_x}{\omega_x \pm \omega}$ (the plus sign corresponds to $\omega_m > \omega_\chi$).

For $\omega > \omega_\chi$, the decay corresponds to the simultaneous excitation of the oscillator and phonons. Since the oscillator has an infinite number of levels and the probability of a transition in which the number of the level increases also increases (neglecting the anharmonicity), a runaway effect takes place, i.e., the stationary distribution is described by a negative temperature $T^* = -T\omega_x/(\omega - \omega_x)$ and cannot be normalized. We shall consider two mechanisms removing the runaway effect: nonlinear friction[8] and anharmonicity. It is found that the current can be limited by an infinitely weak anharmonicity but the limitation of the oscillator energy can occur only when the distribution of the levels (or at least of the high-energy levels) is strongly nonequidistant. The mechanisms limiting the runaway effect do not destroy the population inversion of several lowest levels and, therefore, nonequilibrium local vibrations can amplify light at a resonance frequency.

1. TRANSPORT EQUATION IN A QUASIMONOCHROMATIC FIELD

We shall consider a one-dimensional anharmonic oscillator interacting with continuous-spectrum vibrations representing a thermostat. We shall assume a weak interaction $\Gamma \ll \omega_m, \omega_\chi$. In the simplest case, the Hamiltonian of the system under study in an external field is given by

$$\left.\begin{aligned} H = H_0 + H_{ph} + H_i;\ H_0 = \omega_x a_x^+ a_x + \frac{1}{12} V c_x^4 - f(t)\, c_x;\ c_x = a_x^+ + a_x, \\ f(t) = f_1(t)\exp(-i\omega t) + f_1^*(t)\exp(i\omega t), \\ H_{ph} = \sum_k \omega_k a_k^+ a_k;\ H_i = \sum_k V_{xk} c_x c_k \\ + \frac{1}{2}\sum_{k,k'} V_{xkk'} c_x c_k c_{k'} + \sum_k V_{xxk} c_x^2 c_k;\ c_k = a_k + a_k^+, \end{aligned}\right\} \quad (2)$$

where k labels the continuous spectrum vibrations. The terms $\sim c_\chi^3$ are omitted in H_0 since it is assumed that the equilibrium position of the oscillator represents a center of inversion. The first and second terms in H_i in Eq. (2) lead to a damping and shift of the oscillator frequency due to the decay into one or two phonons; the third term yields maxima in $\mathrm{Im}\,R(\omega)$ for $\omega \sim \omega_\chi \pm \omega_m$ (see Ref. 6) and also a nonlinear friction[8] with a complex coordinate dependence; the latter term corresponds to the decay with the annihilation of two oscillator quanta.

A transport equation based on the Hamiltonian (2) without the field term was derived in Refs. 8-10 and its solution was analyzed in the equilibrium and nonequilibrium cases for an arbitrary ratio of the anharmonicity parameter V and the relaxation rate Γ (but $|V|, \Gamma \ll \omega_m, \omega_\chi$). The method of Ref. 9 can be generalized to an oscillator in a quasimonochromatic field

$$\frac{d\ln|f_1|}{dt} \ll t_c^{-1},\ T\bar{n}(|\omega \pm \omega_x|);\ t_c^{-1} = \min\{\omega,\ \omega_m,\ \omega_x|\omega - \omega_x|\}. \quad (3)$$

We shall assume that $f(t) = 0$ for $t \le t_0$ and the oscillator is in equilibrium with the vibrations belonging to the continuous spectrum. For moderate fields $|Vf^2| \ll t_c^{-3}$, we obtain from Eq. (3) the following result, which holds in the interaction representation:

$$\left.\begin{aligned} \bar{a}_x(t) \simeq a_0(t)e^{-i\omega_x t} + v(t);\ v(t) = \frac{f_1(t)e^{-i\omega t}}{\omega_x - \omega} + \frac{f_1^*(t)e^{i\omega t}}{\omega_x + \omega}; \\ \dot{a}_0(t) = -iV\left[1 + \frac{8\omega_x^2|f_1(t)|^2}{(\omega_x^2 - \omega^2)^2} + a_0^+(t)a_0(t)\right]a_0(t); \\ \bar{A}(t) = S^+(t, t_0)AS(t, t_0);\ S(t, t_0) = T\exp\left[-i\int_{t_0}^{t} d\tau H_0(\tau)\right]. \end{aligned}\right\} \quad (4)$$

The transport equation for the density matrix in the interaction representation $\rho_\chi(t)$ obtained in the second order in H_i has the following form for large times $t - t_0 \gg t_c$:

$$\left.\begin{aligned} \frac{\partial\rho_x}{\partial t} = -\hat{\Gamma}_f\rho_x - \hat{\Gamma}_2\rho_x - iP_f[n_0, \rho_x] - i\frac{V^{(2)}}{2}[n_0^2, \rho_x], \\ \rho_x \equiv \rho_x(t),\ n_0 = n_0(t) = a_0^+a_0, \\ \hat{\Gamma}_f\rho_x = \Gamma_f[(2\bar{n}_f + 1)\{n_0, \rho_x\}_+ + 2\bar{n}_f\rho_x - 2(\bar{n}_f + 1)a_0\rho_x a_0^+ \\ - 2\bar{n}_f a_0^+\rho_x a_0],\ a_0 = a_0(t), \\ \{A, B\}_+ = AB + BA;\ \Gamma_f = \Gamma_f(t) \\ = \Gamma + \frac{16\omega_x^2|f_1(t)|^2}{(\omega_x^2 - \omega^2)^2}(\Gamma_+ + \Gamma_- - \Gamma_{inv}). \end{aligned}\right\} \quad (5)$$

Here, $V^{(2)}$ is the renormalization of the anharmonicity parameter V due to a third term in H_i (such renormalization was obtained in Ref. 9); P_f is a time-dependent frequency shift; for $f = 0$, it was evaluated in Ref. 9; and the field-dependent part of P_f can be obtained by a Kramers–Kronig transformation from $\Gamma_f - \Gamma$. The parameters $\Gamma_\pm$ and Γ_{inv} govern the probability of the stimulated decay:

$$\left.\begin{aligned} \Gamma_\pm = \pi\sum_k V_{xxk}^2\delta(\omega_x \pm \omega - \omega_k);\ \Gamma_{inv} = \pi\sum_k V_{xxk}^2\delta(\omega - \omega_x - \omega_k), \\ \bar{n}_f = \bar{n}_f(t) = \Gamma_f^{-1}\left\{\Gamma\bar{n}(\omega_x) + \frac{16\omega_x^2|f_1(t)|^2}{(\omega_x^2 - \omega^2)^2}\right. \\ \left.\times[\Gamma_+\bar{n}(\omega_x + \omega) + \Gamma_-\bar{n}(\omega_x - \omega) + \Gamma_{inv}(\bar{n}(\omega - \omega_x) + 1)]\right\}. \end{aligned}\right\} \quad (6)$$

The parameter Γ in Eq. (5) is the field-independent decay damping[9] and the operator $\hat{\Gamma}_2\rho_x$ describes a nonlinear friction and is given by (see Ref. 8)

$$\left.\begin{aligned} \hat{\Gamma}_2\rho_x = \Gamma^{(2)}[2\bar{n}(2\omega_x)\{n_0^2 + n_0 + 1, \rho_x\}_+ \\ + \{n_0^2 - n_0, \rho_x\}_+ - 2(\bar{n}(2\omega_x) + 1)a_0^2\rho_x a_0^{+2} \\ - 2\bar{n}(2\omega_x)a_0^{+2}\rho_x a_0^2];\ \Gamma^{(2)} = \pi\sum_k V_{xxk}^2\delta(2\omega_x - \omega_k). \end{aligned}\right\} \quad (7)$$

Equation (5) is derived neglecting the terms $\sim(\bar{n}_f + 1)\cdot\left|V\frac{d\Gamma_f}{d\omega}\right| \ll \Gamma_f$ and $\Gamma_f t_c,\ \Gamma^{(2)} t_c,\ |V|\,t_c,\ |P_f|\,t_c \ll 1$.

It is of interest to compare Eq. (5) and the transport equation for the harmonic oscillator with linear friction discussed in Ref. 4. It follows from Eqs. (1) and (4) that, in the approximation defined by Eq. (3), we obtain $a_0(t) = e^{i\omega_x t}\tilde{D}(t)\bar{a}_x(t)\tilde{D}^+(t)$, where $\tilde{D}(t)$ is defined by Eq. (1), in which a_χ should be replaced by $\tilde{a}_\chi(t)$ and the integration over time is from t_0. Transforming from the interaction representation with the Hamiltonian H_0 to the interaction representation with the Hamiltonian $H_0' = \omega_\chi a_\chi^+ a_\chi$, we find that, up to

rapidly oscillating terms, the transport equation (5) is formally the same as the transport equation obtained in Ref. 4. However, it should be noted that the field-induced decay effects modify the parameters Γ_f and $\bar{n}_f$ in Eq. (5) so that they can deviate considerably from their equilibrium values.

Equation (5) can be also applied to analyze the peaks in the spectral distribution of a nonequilibrium oscillator near ω_χ and $|\pm\omega\pm\omega_\chi|$. The profile of the peaks (linear response to an additional weak field) can be obtained from two-time correlation functions $\langle A(\tau)B(t)\rangle$ at large times $|t-\tau| \gg t_c$. It can be shown (for f = 0, this was demonstrated in Ref. 10) that, with an accuracy up to small terms $\sim\Gamma_f t_c$, we obtain

$$\langle A(\tau)B(t)\rangle \quad \mathrm{Tr}_\chi[\bar{B}(t)\hat{G}(t,\tau;\rho_\chi(\tau)\bar{A}(\tau))],\ t-\tau\gg t_c, \tag{8}$$

where Tr_χ represents the trace taken over the wave functions of an isolated oscillator. The operator $\hat{G}\{t,\tau;\bar{C}(\tau)\}$ describes the relaxation of the oscillator and satisfies the differential (with respect to t) equation (5) with the initial condition $\hat{G}\{\tau,\tau;\bar{C}(\tau)\}=\bar{C}(\tau)$, where

$$\rho_\chi(t)=\hat{G}\{t,t_0;Z^{-1}\exp[-\lambda H_0(t_0)]\};\ Z=\mathrm{Tr}_\chi\exp[-\lambda H_0(t_0)]. \tag{9}$$

2. SOLUTION OF THE TRANSPORT EQUATION FOR A MONOCHROMATIC FIELD

For a monochromatic field, we obtain $|f_1(t)| = \text{const}$ and it follows from Eqs. (4) and (5) that the parameters in our transport equation cease to depend on time. During a time $\Delta t \gtrsim \Gamma_f^{-1}$, a steady-state distribution of the oscillator with respect to the eigenfunctions of the operator n_0 is reached [it follows from Eq. (4) that $dn_0/dt = 0$]. We shall first discuss this distribution for $\Gamma^{(2)} = 0$. The steady-state solution of Eq. (5) is then given by the Boltzmann distribution

$$\rho_\chi(t)=\rho_\chi=(\bar{n}_f+1)^{-1}\exp(-\omega_\chi n_0/T^*),\ T^*=\omega_\chi\left[\ln\frac{\bar{n}_f+1}{\bar{n}_f}\right]^{-1}. \tag{10}$$

It follows from Eq. (4) that the density matrix in the Schrödinger representation is a shifted Boltzmann distribution and Eq. (10) has formally the same form as Eq. (1). However, the effective temperature T^* in Eq. (10) can be either much higher or much lower than the thermostat temperature. In particular, for $\Gamma_f \approx 16\omega_\chi^2|f_1|^2\Gamma_\pm/(\omega_\chi^2-\omega^2)^2$, we obtain $T^*\approx T\omega_\chi/(\omega_\chi\pm\omega)$.

In weak fields, we find that

$$T^*\approx T+\frac{16\omega_\chi^2|f_1|^2}{(\omega_\chi^2-\omega^2)^2}\frac{T^2}{\Gamma\omega_\chi\bar{n}(\omega_\chi)[\bar{n}(\omega_\chi)+1]}\{\Gamma_+[\bar{n}(\omega_\chi+\omega)-\bar{n}(\omega_\chi)]$$
$$+\Gamma_-[\bar{n}(\omega_\chi-\omega)-\bar{n}(\omega_\chi)]+\Gamma_{1\mathrm{inv}}[\bar{n}(\omega-\omega_\chi)+\bar{n}(\omega_\chi)+1]\},\ |\Gamma_f-\Gamma|\ll\Gamma. \tag{11}$$

It follows from Eq. (11) that the effective temperature changes rapidly also in weak fields provided the equilibrium Planck number of the oscillator is small, $\bar{n}(\omega_\chi) \ll 1$, $\bar{n}(|\omega_\chi-\omega|) \gg \bar{n}(\omega_\chi)$ and a decay $\omega_k=|\omega-\omega_\chi|$ can occur. It should be noted that, when Eq. (10) is satisfied, the steady-state oscillator current $\langle c_\chi(t)\rangle \approx {}_\chi[\bar{c}_\chi(t)\rho_\chi(t)]=\frac{4\omega_\chi}{\omega_\chi^2-\omega^2}\mathrm{Re}[f_1e^{-i\omega t}]$ has nonzero terms only at the field frequency.

Equation (10) ceases to be valid when the dominant relaxation mechanism is the decay of the field quanta accompanied by the creation of a phonon and excitation of the oscillator since $\Gamma_f < 0$, $\bar{n}_f \leq -1$, and $T^* < 0$ and the distribution (10) can no longer be normalized. For $\Gamma_f < 0$ ($\Gamma^{(2)} = 0$), a runaway effect for the oscillator takes place. For example, the average occupation number obtained from Eq. (5) is given by $d\langle\bar{n}_0\rangle/dt=-\Gamma_f(\langle n_0\rangle-\bar{n}_f)$; for $\Gamma_f < 0$ [it follows from Eq. (6) that $\Gamma_f\bar{n}_f > 0$], we obtain $\langle n_0\rangle \sim \exp(|\Gamma_f|t)\to\infty$ for $t\to\infty$.

To analyze the runaway effect and to calculate the correlation functions defined by Eq. (8), it is convenient to transform from the operator equation (5) to difference equations and take the matrix elements with respect to a complete orthonormal system of functions $|m)_\tau$:

$$|m)_\tau=(m!)^{-1/2}[a_\tau^+(\tau)e^{i\omega_\chi\tau}]^m|0)_\tau;\ |0)_\tau=\bar{D}(\tau)S^+(\tau,t_0)|0);$$
$$_\tau(m'|n_0(t)|m)_\tau=m\delta_{m',m}. \tag{12}$$

Here, $|0)$ is the wave function of the oscillator ground state at the time t_0 [$|m)_\tau$ are not eigenfunctions of the Hamiltonian H_0]. The parameter τ in Eq. (12) is arbitrary but it is convenient in our discussion of the relaxation to assume that τ is a time at which the initial condition for our transport equation is specified. For such a choice of τ, it follows from Eq. (5) that $\hat{G}(t,\tau)$ depends only to $t-\tau$.

The system of differential–difference equations for $\rho_{mn}(t) = {}_\tau(m|\rho_\chi(t)|n)_\tau$ corresponding to $|f_1(t\geq\tau)| = \text{const}$ has the same form as the system of equations for ρ_{mn} obtained in Refs. 8 and 10 for f = 0. The method of generating functions was used in Eq. (10) to solve the aforementioned system; the obtained solution [Eqs. (11)–(13) of Ref. 10] corresponding to large times remains valid even for $\Gamma_f < 0$. The evolution of the matrix elements can be easily studied in the case $\bar{n}_f = -1$ and $T^* = -0$. Let us assume that $\rho_{00} = 1$ for $t = \tau$. As a result, we obtain

$$\rho_{nn}(t)=\exp[-2|\Gamma_f|(t-\tau)](1-e^{-2|\Gamma_f|(t-\tau)})^n,\ t\geq\tau. \tag{13}$$

It follows from Eq. (13) that $\rho_{nn}(t)\to 0$ for $t\to\infty$ and an arbitrary finite n but, as $t-\tau$ increases, states with higher and higher numbers n are excited (the runaway effect) and the quantity $\langle n_0(t)\rangle=\sum_n n\rho_{nn}(t)$ grows. The internal anharmonicity of the oscillator considered in the approximation $\langle n_0\rangle\left|V\frac{d\Gamma_f}{d\omega}\right|\ll\Gamma_f$ does not influence the diagonal elements of ρ_χ but is very important for the nondiagonal elements. For example, for $\bar{n}_f = -1$ and $\Gamma_f < 0$, the quantity $\rho_{n+1n}(t)$ (this matrix element governs the oscillator current at a frequency ω_χ) can be obtained from Eq. (5) under the assumption $\rho_{10}(\tau)=\alpha,\ \rho_{n+1n}(\tau)|_{n>0}=0$ in the following form:

$$\rho_{n+1n}(t)=\alpha\left[1+i\frac{V+V^{(2)}}{2|\Gamma_f|}\right]^{-n}\sqrt{n+1}$$
$$\exp\{-[3|\Gamma_f|+i(P_f-Vn+V^{(2)}/2)](t-\tau)\}$$
$$\times(1-\exp[-(2|\Gamma_f|+iV+iV^{(2)})(t-\tau)])^n. \tag{14}$$

It follows from Eq. (14) that $\langle a_0(t)\rangle \propto \sum_n\sqrt{n+1}\rho_{n+1n}(t)\exp[-iVn(t-\tau)] \propto \exp[-3|\Gamma_f|(t-\tau)]$ for $t-\tau\to\infty$ provided $V + V^{(2)} \neq 0$; for $V^{(2)} + V = 0$, a runaway of the current takes place[1]: $\langle a_0(t)\rangle \propto \exp[|\Gamma_f|(t-\tau)]$. The damping of $\langle a_0(t)\rangle$ due to a nonequidistant distribution of the levels can be explained as follows: For a nonequidistant distribution of the levels, there are always levels with such high numbers that the transitions between these levels have frequencies that differ considerably from the transition $|0)_\tau \to |1)_\tau$; the probability of the excitation of such transitions is very low, $\left|1+i\frac{V+V^{(2)}}{2|\Gamma_f|}\right|^{-N}\ll 1$, $N\gg 1$. For a harmonic oscillator, all

transitions occur at the same frequency and, therefore, they interfere. As a result, only the relative contribution of a single transition can be defined.[8] Because of interference, the total current grows exponentially as a function of time.

There are several mechanisms restricting the oscillator runaway. The simplest mechanism is related to a nonequidistant distribution of the excited levels. If, for some level N_0, the transition frequency $\omega(n)$ between the n-th and (n + 1)-th levels ($n \geq N_0$) deviates to such an extent from $\omega(0) \approx \omega_\chi$ that the decay $\omega = \omega_k + \omega(n)$ cannot occur, the runaway effect does not arise. The aforementioned model cannot be described by the transport equation (5) since the damping operator $\hat{\Gamma}_f \rho_x$ corresponding to this mechanism is more complex and, in general, an oscillator temperature cannot be introduced. However, assuming that, for $n \leq N_0$, decay effects $\omega = \omega_k + \omega\ \omega(n)$ dominate, it is possible to introduce an oscillator temperature in the corresponding energy range provided $\omega_\chi > T \gg \omega(N_0) - \omega_\chi$, i.e.,

$$\left.\begin{aligned} &\rho_{nn} = A \exp(-\omega_\chi n/T^*),\ n \leq N_0;\ \rho_{nn} = 0,\ n > N_0; \\ &A = \frac{1 - \exp(-\omega_\chi/T^*)}{1 - \exp[-(N_0+1)\omega_\chi/T^*]}, \\ &T^* \approx -\omega_\chi T/(\omega - \omega_\chi),\ \Gamma \ll \Gamma_{inv} \end{aligned}\right\} \quad (15)$$

[Eq. (15) can be obtained from Eq. (5) for arbitrary $\Gamma/\Gamma_{inv} < 1$ provided the weighted phonon density of states is rectangular]. The population of the levels defined by Eq. (15) increases monotonically up to a level N_0 and there decays rapidly to zero. For a real anharmonic oscillator, such a sharp dependence is smeared. For low-energy levels, the distribution is only weakly nonequidistant and it follows from Eqs. (5) and (10) that it is possible to define a negative temperature for such levels provided the inverting pumping is strong. For higher levels, the contribution of the pumping decreases and then becomes small compared with the contribution of other relaxation mechanisms. As a result, the population of the levels is a function with a wide maximum.

Another mechanism contained in our model which can also limit the runaway effect is nonlinear friction defined by Eq. (7). It follows from Eq. (7) that nonlinear friction increases rapidly with increasing oscillator energy and, therefore, even for small $\Gamma^{(2)}/\Gamma_f$ it becomes important for high levels. To analyze the contribution of nonlinear friction, it is convenient to transform from the difference equation for ρ_{nn} to a differential equation for a generating function: $\varphi(s) = \sum_{n=0}^{\infty} \rho_{nn} s^n$

$$\varphi'(s^2 - e^{2\omega_\chi/T}) + 4s\varphi' + 2\varphi = -\frac{\Gamma_f \bar{n}_f}{\Gamma^{(2)} \bar{n}(2\omega_\chi)(s+1)} \times \{\varphi'[s - \exp(\omega_\chi/T^*)] + \varphi\},\ \varphi(1) = 1. \quad (16)$$

The requirement that the function $\varphi(s)$ should be analytic in a unit circle represents the second boundary condition to Eq. (16). We shall illustrate the application of this condition in our calculation of ρ_{nn} for $\Gamma_f \ll \Gamma^{(2)}$. In the zeroth approximation in Γ_f, we obtain

$$\varphi(s) = \frac{\rho_{00} + s\rho_{11}}{1 - s^2 e^{-2\omega_\chi/T}},\ \rho_{00} + \rho_{11} = 1 - e^{-2\omega_\chi/T},$$
$$\rho_{nn} = e^{-2\omega_\chi/T} \rho_{n-1\,n-2},\ \Gamma^{(2)} \gg \Gamma_f. \quad (17)$$

To calculate ρ_{00} and ρ_{11}, we note that the right-hand side of Eq. (16) remains finite for $\Gamma_f \neq 0$ at the point $s = -1$ provided the expression in braces vanishes at this point. This yields

$$\rho_{11}/\rho_{00} = [1 + e^{-2\omega_\chi/T}(1 + 2e^{\omega_\chi/T^*})]/[2 + e^{\omega_\chi/T^*}(1 + e^{-2\omega_\chi/T})]. \quad (17a)$$

For $\Gamma^{(2)} \gg \Gamma_f$, it follows from Eqs. (17) and (17a) that, irrespective of the ratio T^*/T and of the sign of T^*, population inversion does not occur, $\rho_{11} < \rho_{00}$.

For $\Gamma^{(2)} \ll \Gamma_f$, population inversion can occur for $T^* < 0$. The standard perturbation theory in $\Gamma^{(2)}/\Gamma_f$ cannot be used to analyze the limitation of the runaway effect by nonlinear friction. However, nonlinear friction can be taken into account consistently in the important limiting case of low temperatures $\exp(-2\omega_\chi/T) \ll 1$ when Eq. (16) can be reduced to a degenerate hypergeometric equation. For $\Gamma^{(2)} < \Gamma_f(2\bar{n}_f + 1)$, the only analytic solution of this equation is given by

$$\varphi(s) = {}_1F_1\left(1,\ \frac{\Gamma_f(2\bar{n}_f+1)}{\Gamma^{(2)}},\ \frac{\Gamma_f \bar{n}_f}{\Gamma^{(2)}}(s+1)\right)/{}_1F_1\left(1,\ \frac{\Gamma_f(2\bar{n}_f+1)}{\Gamma^{(2)}},\ \frac{2\Gamma_f \bar{n}_f}{\Gamma^{(2)}}\right), \quad (18)$$

where ${}_1F_1$ is the degenerate hypergeometric function.[12] Using Eq. (18), we can easily verify that $\rho_{00} < \rho_{11}$ is satisfied for weak nonlinear friction provided $T^* < 0$, i.e., population inversion is obtained.

The nonlinear friction defined by Eq. (7) can be the dominant mechanism removing the runaway of low-frequency quasilocal vibrations since the parameter $\Gamma^{(2)}$ is relatively large since the phonon density of states at the frequency $2\omega_\chi$ is higher than the density of states at the frequency ω_χ. For local vibrations, we obtain $\Gamma^{(2)} = 0$. However, for $\omega_m < \omega_\chi < 2\omega_m$, the nonlinear friction of local vibrations can be due to the field and is obtained if the terms $\sum_k V_{xxk} c_x^2 c_k$ in H_1 in Eq. (2) are taken into account. For higher frequency vibrations, the principal mechanism limiting the runaway effect is a strong anharmonicitiy which manifests itself for such vibrations.

It should be noted that, for a strong field satisfying $\Gamma_f \gg \Gamma$, the transport equation (5) has a quasistationary solution defined by Eq. (10) even for a nonmonochromatic field.

3. INTERACTION OF NONEQUILIBRIUM LOCAL VIBRATIONS WITH LIGHT

A convenient method of study of the states of nonequilibrium oscillators is the investigation of their absorption (amplification) and luminescence spectra. We shall discuss the profile of peaks near resonance frequencies in the spectrum of local vibrations located in a nonresonant monochromatic field under steady-state conditions, i.e., when the susceptibilities averaged over the period ω^{-1} are independent of time. The polarizability of the oscillator dipole moment $M = \mu c_\chi$ is determined (see Ref. 10) by the Fourier transform of the correlation function $\langle [c_x(t),\ c_\chi(\tau)] \rangle$. Under steady-state conditions, we obtain $\langle [c_x(t),\ c_x(\tau)] \rangle = \langle [c_\chi(t-\tau),\ c_\chi(0)] \rangle$. It follows from Eq. (4) that, with an accuracy up to small corrections $\sim \Gamma_f t_c$, the profile of the absorption spectrum near the normal frequency of the oscillator is described by

$$Q_0(\Omega) = \mathrm{Re} \int_0^\infty dt \exp(i\Omega t) \langle [c_x(t),\ c_x(0)] \rangle \simeq \mathrm{Re} \int_0^\infty dt \exp(i\Omega t)\, Q_0(t);$$

$$Q_0(t) = \langle [a_x(t),\ a_x^+(0)] \rangle$$

$$\approx e^{-i\omega_x t}\,\mathrm{Tr}_x\,[a_0(t)\,\hat{G}\,(t,\ 0;\ [a_0^+(0),\ \rho_x(0)])],\ \Omega \sim \omega_x. \qquad (19)$$

Using Eqs. (5)-(7) and (12), we can write $Q_0(\Omega)$ as a set of nonoverlapping lines corresponding to the transitions between neighboring levels of the oscillator induced by resonant radiation. The system of equations describing the interference of such lines has the same form as in the equilibrium case.[8, 9] However, in the nonequilibrium case, the parameters Γ_f and T^* and also the transition probabilities that are proportional to the differences between the populations of the levels are field dependent.

For decay effects $\omega_k = \omega_\chi \pm \omega$, the broadening Γ_f is an increasing function of the field. However, for $\bar{n}(\omega_\chi) \gg 1$ and $|V| > \Gamma$, the total width $Q_0(\Omega)$ can decrease in a cooling field ($\Gamma_\perp \gg \Gamma_-$, Γ_{inv}), since transitions from fewer levels contribute to the formation of $Q_0(\Omega)$ and the effect of a nonequidistant distribution of levels is weaker. For $|V| \ll \Gamma_f$, a reduction of the half-width Γ_f of $Q_0(\Omega)$ occurs in a field provided $\Gamma_{inv} \gg \Gamma_\pm$. The effective oscillator temperature increases with decreasing Γ_f and, for $\Gamma_f \sim \bar{n}_f|V|$, the distribution $Q_0(\Omega)$ broadens due to a nonequidistant distribution of the levels. For $\Gamma_f < 0$ and $\Gamma_f(2\bar{n}_f + 1) > \Gamma^{(2)}$, a nonequilibrium oscillator amplifies the light in a certain interval of frequencies. It follows from Eq. (19) that the absorption of light takes place in the whole spectrum

$$\int d\Omega Q_0(\Omega) \simeq \pi \langle [a_x(0),\ a_x^+(0)] \rangle = \pi.$$

Therefore, local vibrations with a strong anharmonicity $|V| \gg \Gamma_f$ lead to optimum amplification. The corresponding spectrum $Q_0(\Omega)$ represents a set of weakly overlapping nearly equidistant lines. If the intensity of the n-th line, which is proportional to $(n+1)(\rho_{nn} - \rho_{n+1\,n+1})$, is negative, amplification takes place in the corresponding frequency interval.

It was shown in Ref. 13 that a cubic anharmonicity $\tilde{V}c_\chi^3$ and a nonlinear dependence of the dipole moment of local vibrations on the displacement leads to a resonance absorption of light at twice the frequency of the oscillator. This result holds also in nonequilibrium under steady-state conditions. The profile of the peak near the doubled frequency is governed by the Fourier transform of the correlation function

$$Q_1(t) = \mathrm{Tr}_x\,[\tilde{c}_x^2\,(t)\,\hat{G}\,(t,\ 0;\ [\tilde{c}_x^2(0),\ \rho_x(0)])], \qquad (20)$$

where the quantity $\tilde{c}_\chi^2(t)$ should be replaced by $a_0^2(t)\exp(-2i\omega_\chi t)$ and the quantity $\tilde{c}_\chi^2(0)$ should be replaced by $[a_0^+(0)]^2$. However, Eq. (20) can be used to study the profile of the peak not only at twice the oscillator frequency (the corresponding calculations for the equilibrium case were made in Ref. 14) but also near the Raman spectrum $|\pm\omega \pm \omega_x|$. It is only necessary to take into account in $\tilde{c}_\chi^2$ the cross terms $(v + v^*)(a_0 + a_0^+)$. For $\omega_\chi \pm \omega > 0$, the term in $Q_1(t)$ which governs the peak at a frequency $\Omega \sim \omega_x \pm \omega_\chi$ differs from $Q_0(t)$ only by an additional factor $16\omega_x^2|f_1|^2(\omega_x^2 - \omega^2)^{-2} \times e^{\mp i\omega t}$ and, therefore, peaks at the frequencies ω_χ and $\omega_\chi \pm \omega$ have the same profiles. For $\omega > \omega_\chi$, the peak at a frequency $\Omega \sim \omega - \omega_\chi$ is governed by the correlation function

$$\tilde{Q}_1(t) = \frac{16\omega_x^2}{(\omega^2 - \omega_x^2)^2}|f_1|^2 e^{-i(\omega-\omega_x)t}\,\mathrm{Tr}_x\,[a_0^+(t)\,\hat{G}\,(t,\ 0;\ [a_0(0),\ \rho_x(0)])]. \qquad (21)$$

A comparison of Eqs. (21) and (19) indicates that the functions $\frac{16\omega_x^2|f_1|^2}{(\omega^2-\omega_x^2)^2} Q_0(x+\omega)$ and $\tilde{Q}_1(x+\omega-\omega_x) \times (|x| \ll \omega,\ \omega_x)$ exhibit an inverse symmetry with respect to x = 0. For optical pumping, the regions $\tilde{Q}_1(\Omega)$ ($\Omega \sim \omega - \omega_\chi$) of amplification (for $\Gamma_f > 0$, this includes the whole peak region) correspond to the stimulated Raman scattering of light.

The Raman scattering of light interacting with local vibrations and the stimulated Raman scattering of light are due to the anharmonicity and nonlinear polarizability of the oscillator. The Raman scattering and luminescence at the normal frequency can be calculated as in Ref. 10 provided the interaction with the quantized electromagnetic field in the crystal is taken into account. For $\Gamma_f > 0$ and $\Gamma^{(2)} = 0$, the profile of the luminescence peaks near ω_χ and $\omega - \omega_\chi$ is governed by

$$W_0(\Omega) = [\bar{n}_f - n(\Omega)]\,Q_0(\Omega),\ \Omega \sim \omega_x;\ \tilde{W}_1(\Omega)$$
$$= -[\bar{n}_f + n(\Omega) + 1]\,\tilde{Q}_1(\Omega),\ \Omega \sim \omega - \omega_x\ (\tilde{W}_1 > 0). \qquad (22)$$

The oscillator emits light at the normal frequency and at frequencies $\omega_x \pm \omega > 0$, provided its effective temperature is higher than the temperature of the medium. At a frequency $\omega - \omega_\chi$ (for $\omega - \omega_\chi > 0$), the emission of light (Raman scattering) takes place for arbitrary $T^*/T > 0$. For $\Gamma_f < 0$, Eq. (22) is no longer applicable; at frequencies $\Omega \sim \omega - \omega_\chi$ there are frequency intervals in which the stimulated Raman scattering does not manifest itself, i.e., $\tilde{Q}_1(\Omega) > 0$ and, for large $\bar{n}(\Omega)$, no luminescence occurs.

Equation (20) can be also applied to the absorption of nonresonant phonons by a nonequilibrium oscillator [the absorption coefficient is proportional to $\sum_k V_{xxk}^2 \delta(\Omega - \omega_k) Q_1(\Omega)$]. It follows from Eqs. (20) and (22) that the oscillator can emit phonons spontaneously at frequencies $\omega_k \sim \omega_x \pm \omega$ for $\bar{n}(\omega_k) < \bar{n}_f$ but no amplification takes place. Phonons at frequencies $\omega_k \sim \omega - \omega_\chi$ are not only spontaneously emitted but it follows from Eq. (21) that coherent amplification of the phonons in a field takes place.

When the pumping of localized impurity vibrations is due to a nonresonant light, the largest effect is obtained when the frequency of the light lies at the maximum of one of the side absorption bands (absorption with the participation of a phonon). The aforementioned bands are due to the anharmonic terms $c_\chi^2 c_k$ in Eq. (2) and due to a nonlinear polarizability of local vibrations[7] depending on the phonon coordinates. The nonlinear polarizability leads to a term $f(t)\sum_k m_{xk} c_x c_k$ in the Hamiltonian defined by Eq. (2). The coefficients $m_{\chi k}$ lead to a renormalization of $V_{\chi\chi k}$ in the expressions for the induced decay parameters $\Gamma_\pm$, Γ_{inv}: $V_{xxk} \to V_{xxk} + \frac{\omega_x^2 - \omega^2}{4\omega_x} m_{xk}$.

Fields that are required for the rate of the induced decay to be of the order of the spontaneous relaxation rate can be estimated from the measured ratio of the absorption at the maximum of the peak α_m to the absorption in the side band $\alpha_{s.b.}(\omega)$: $|f|^2 \sim \Gamma\Gamma_0\omega\alpha_m/(\omega_x\alpha_{s.b.})$, where Γ_0 is the half-width of the peak. The estimated fields are weaker than 10^5 V/cm. The amplification of light and phonons at the difference frequency $\omega - \omega_\chi$ can manifest itself in weaker fields; the threshold for such an amplification is governed by the lattice absorption. To obtain strong heating of local vibrations or cooling of quasilocal vibrations,

it is necessary to select high-frequency vibrations in the former case and low-frequency vibrations in the latter case so that Γ is small. Uniform heating of high-frequency local vibrations up to high occupation numbers $\bar{n}(\omega_\varkappa) \gtrsim 1$ is often impossible.

Our results obtained for a one-dimensional local vibration can be generalized to the case of several anharmonically interacting local vibrations. In the nondegenerate case when different local vibrations have different frequencies and the field frequency is not equal to the difference or combination frequency of local vibrations, such a generalization is trivial: the interaction $c_\varkappa c_{\varkappa'} c_k$ results in a situation in which the field acting on an oscillator $\varkappa$ induces the decay of the oscillator $\varkappa'$ and vice versa. Such decay effects are analogous to those that have been already described. However, for degenerate vibrations (for example, local vibrations of cubic symmetry), the situation is quite different. In general, for an arbitrary orientation of the field, the problem in question cannot be solved and, in particular, an effective temperature of local vibrations cannot be defined. However, in the important special case of high-frequency local vibrations, an effective temperature can be introduced approximately. In fact, it is only necessary to note that, for such local vibrations, there is a fast relaxation mechanism due to the quasielastic scattering of phonons from local vibrations. Such a scattering leads to a levelling of the occupation numbers of different local vibration states with approximately equal energies (with the same principal quantum number).

Large effects can occur in relatively weak fields if the field frequency is close to the difference of the frequencies of two anharmonically interacting ($V_{\varkappa\varkappa\varkappa_1} c_\varkappa^2 c_{\varkappa_1}$) local vibrations provided one of these vibrations is rapidly relaxing,[2)] $\Gamma_{\varkappa_1} \gg \Gamma_\varkappa$, $|V_{\varkappa\varkappa\varkappa_1}|$. For $\Gamma_{\varkappa_1}^2 \gg V_{\varkappa\varkappa\varkappa_1}^2 |f_1|^2 \omega_\varkappa^2/(\omega_\varkappa^2 - \omega^2)^2$ the relaxation of an oscillator $\varkappa$ is then described by Eq. (5) in which $\Gamma_\pm$, Γ_{inv} should be replaced by a considerably larger parameter $V_{\varkappa\varkappa\varkappa_1}^2/\Gamma_{\varkappa_1}$. The resonance method considered is particularly important for the cooling of low-frequency quasilocal vibrations. This is due to the fact that, for the induced relaxation to phonons, it is difficult to satisfy the necessary inequality $\Gamma_+ \gg \Gamma_{\text{inv}}$ for such vibrations. For a resonance with high-frequency local vibrations, the latter inequality can be easily satisfied provided $\omega_\varkappa \gg \Gamma_{\varkappa_1}$.

[1)] It was shown in Ref. 11 that the runaway effect which occurs in semiconductors for the scattering of electrons from polar optical phonons can be limited by a finite width of the forbidden band.

[2)] As an example of such a system, we may quote high-frequency (intramolecular) local vibrations of an impurity molecule interacting with relatively low-frequency (in particular, libration) vibrations, whose damping is strong.

[1] M. A. Ivanov, L. B. Kvashnina, and M. A. Krivoglaz, Fiz. Tverd. Tela (Leningrad) 7, 2047 (1965) [Sov. Phys. Solid State 7, 1652 (1966)].
[2] N. N. Bogolyubov, Statistical Methods in Mathematical Physics [in Russian], Akad. Nauk SSSR, Kiev (1945).
[3] J. Schwinger, J. Math. Phys. (N. Y.) 2, 407 (1961).
[4] B. Ya. Zel'dovich, A. M. Perelomov, and V. S. Popov, Zh. Eksp. Teor. Fiz. 55, 589 (1968); 57, 196 (1969) [Sov. Phys. JETP 30, 111 (1970)].
[5] M. Lax, Fluctuations and Coherence Phenomena in Classical and Quantum Physics [Russian translation], Mir, Moscow (1974).
[6] M. I. Dykman and M. A. Krivoglaz, Ukr. Fiz. Zh. 19, 124 (1974).
[7] A. A. Maradudin, "Theoretical and experimental aspects of the effects of point defects and disorder on the vibrations of crystals," Solid State Phys. 18, 273 (1966); 19, 1 (1966).
[8] M. I. Dykman and M. A. Krivoglaz, Phys. Status Solidi B 68, 111 (1975).
[9] M. I. Dykman and M. A. Krivoglaz, Zh. Eksp. Teor. Fiz. 64, 993 (1973) [Sov. Phys. JETP 37, 506 (1973)].
[10] M. I. Dykman, Zh. Eksp. Teor. Fiz. 68, 2082 (1975).
[11] M. I. Dykman and P. M. Tomchuk, Fiz. Tverd. Tela (Leningrad) 8, 1343 (1966) [Sov. Phys. Solid State 8, 1075 (1966)].
[12] I. S. Gradshteyn and I. M. Ryzhik, Table of Integrals, Series and Products, Academic Press, New York (1965).
[13] M. A. Krivoglaz and I. P. Pinkevich, Opt. Spektrosk. 23, 571 (1967).
[14] M. I. Dykman, Fiz. Tverd. Tela (Leningrad) 15, 1075 (1973) [Sov. Phys. Solid State 15, 735 (1973)].

Translated by D. Mathon