Profiles of no-phonon lines of impurity centers interacting with local or quasilocal vibrations

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The spectral distributions of no-phonon lines of impurity centers in crystals are studied. Their modulation broadening due to interaction of impurity electrons with local or quasilocal vibrations is considered. Using the method employed in the theory of nonlinear oscillators interacting with a medium, the spectral distributions are calculated in the general case of an arbitrary ratio of the parameters governing interaction of electrons with localized modes V_χ and their damping Γ_χ (but assuming $|V_\chi|, \Gamma_\chi \ll \omega_\chi$, where ω_χ is the frequency of a mode χ). The spectral distributions are expressed as integrals of elementary functions. The width, profile, and fine structure of the corresponding strongly non-Lorentzian distribution of a no-phonon line are investigated. Simple analytic expressions for the spectral distributions are obtained in the limits of large and small ratios $|V_\chi|/\Gamma_\chi$ and at high and low temperatures. The intermediate cases are studied numerically.

There are very narrow no-phonon (NP) lines in electron–vibration absorption or emission optical spectra of impurity centers in crystals. It was shown in Ref. 1 that such lines exist even for quite a strong electron–phonon interaction. The NP lines are not broadened by this interaction in the situation when only the interaction of electrons with crystal lattice vibrations which is linear in the phonon operators is important. This result holds in the harmonic approximation and in the absence of inhomogeneous broadening and nonradiative transitions. The width of an NP line is then determined by its natural width. When the quadratic interaction is included, it is found that the modulation of the frequency of an electron transition due to vibrations of atoms leads at finite temperatures to a modulation broadening of the NP lines.[2-7]

It was shown in Refs. 4 and 5 that the interaction of electrons with local or quasilocal vibrations which may occur near impurity centers can lead to a strong modulation broadening of the NP lines (provided such vibrations are excited at the temperatures under study). Such broadening can be much larger than the broadening due to the interaction with vibrations belonging to the continuous spectrum (phonons). In fact, the modulation broadening is determined by the effective density of vibration states $g(\omega)$ which is of the order of Γ_κ^{-1} for a localized mode κ, where Γ_κ is the damping of the mode. On the other hand, the phonon density of states is given by $g(\omega) \propto 1/\Delta\omega$, where $\Delta\omega$ is the width of the phonon band. The damping Γ_κ can be several orders of magnitude smaller than $\Delta\omega$.

The modulation broadening due to local vibrations κ occurs since such local vibrations shift the frequencies of electron transitions by $V_\kappa n_\kappa$ proportional to the occupation number of the mode n_κ (for a small coupling constant V_κ). Assuming that there is no relaxation of vibrations during a characteristic time of the order of the reciprocal width of the NP line γ^{-1}, i.e., for $\gamma \gg \Gamma_\kappa$, the occupation number n_κ remains fixed during such times but the occupation numbers of different centers are different (due to fluctuations). As a result, a set of lines corresponding to pure electron transitions and separated from one another by $|V_\kappa|$ is observed. Such a set of lines corresponds to an NP line with a well-defined fine structure.

Let us now assume that relaxation effects lead to quantum transitions between the levels of localized vibrations in a time $\sim\gamma^{-1}$, i.e., the condition $\gamma \lesssim \Gamma_\kappa$ is satisfied. It follows that averaging of the occupation numbers n_κ occurs during such time for each center and the effect of their fluctuation scatter on the spectrum of an NP line is weakened. It follows that the fine structure of an NP line becomes less pronounced for large Γ_κ and disappears eventually; its width γ for $\Gamma_\kappa \gg |V_\kappa|$ is much smaller than $|V_\kappa|$. This dynamic narrowing of the NP line is analogous to narrowing of the line in the spectrum of systems with a random modulation of the frequency studied phenomenologically in Refs. 8 and 9.

The modulation broadening of an NP line due to interaction with local or quasilocal vibrations was studied in Refs. 4 and 5 in the limiting cases $\Gamma_\kappa \gg \gamma$ and $\Gamma_\kappa = 0$. In view of recent progress in the selective spectroscopy methods which can detect a homogeneously broadened NP line and determine both its homogeneous broadening γ and profile, and also since NP lines have been studied extensively, it is of interest to investigate the profile of an NP line (including its fine structure) in the general case of an arbitrary ratio of γ to Γ_κ.

The proximity (but not equality) of the Bohr frequencies of electron transitions corresponding to different occupation numbers of local vibrations n_κ is a special feature of the problem. Complex interference effects arise in such a "nearly degenerate" many-level system and the usual methods of calculation of the line profile (of Weisskopf–Wigner type) are not applicable and new methods need to be developed. Such methods have been develoepd to study the spectra of nonlinear classical[10] and quantum[11,12] oscillators. We shall apply these new methods to study the modulations of NP lines can be expressed explicitly as an integral of an elementary function and it is possible to discuss in detail the dependences of the line profile and of the width of an NP line on the parameters V_κ and Γ_κ for an arbitrary ratio of these two parameters.

Integral representations for the cross section of the impurity absorption of light in the NP line region $\sigma(\omega)$ are derived in Sec. 1 [the intensity of emission of light from impurity centers is also proportional to $\sigma(\omega)$ in this region and the propor-

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tionality factor is well known]. The profile and fine structure of an NP line and their dependences on the parameters of the electron–phonon interaction, damping of modes Γ_κ, and on temperature are discussed in Sec. 2.

1. SPECTRAL DISTRIBUTION IN THE NO-PHONON LINE REGION

We shall consider an optical transition between the nondegenerate electron levels of an impurity center (the separation between the levels is assumed to be large compared with γ in the case of multiplets); we can then describe the centers using a two-level model. It is also assumed that localized vibrations κ are nondegenerate. We shall consider only systems in which the electron–phonon interaction is weak, which means that only the lowest terms in the interaction constant can be considered in the calculation of the width γ and of the other parameters of the spectrum.

The Hamiltonian of a two-level center interacting with lattice vibrations can be written in the form

$$\left.\begin{aligned} H=H_0+H_i'+H_i'', \quad H_0=\sum_s E_s b_s^+ b_s+\sum_\varkappa \omega_\varkappa a_\varkappa^+ a_\varkappa+\sum_k \omega_k a_k^+ a_k, \\ H_i'=\sum_{\varkappa s} V_{\varkappa ss} b_s^+ b_s a_\varkappa^+ a_\varkappa, \quad H_i''=\sum_\varkappa (a_\varkappa+a_\varkappa^+) h_\varkappa \end{aligned}\right\} \quad (1)$$

Here, $\hbar = 1$; b_s^+ and b_s are the creation and annihilation operators of an electron in the states s (s = 1, 2); a_κ^+, a_κ and a_k^+, a_k are the creation and annihilation operators of localized vibrations κ and of phonons k; H_i' is the diagonal part of the Hamiltonian describing the interaction of electrons with localized modes; H_i'' is the part of the Hamiltonian describing their interaction with phonons which is linear in the coordinates of such modes

$$h_\varkappa \equiv h_\varkappa(a_k, a_k^+)=\sum_k V_{\varkappa k} a_k+\sum_{kk'} V_{\varkappa kk'} a_k^+ a_{k'}+\sum_{kk'} V'_{\varkappa kk'} a_k a_{k'}+\text{H.c.}+\ldots .$$

The terms off-diagonal in b_s^+, b_s neglected in H_i' of Eq. (1) (linear and quadratic in the vibration coordinates) lead to a renormalization of the transition frequency

$$\omega_0=E_2-E_1 \quad (2)$$

and of the coefficients $V_{\kappa ss}$, and also to a decay broadening of the NP line γ_d in the situation when ω_0 lies in the region of normal or combination frequencies of vibrations.[5] The terms diagonal in b_s^+, b_s which do not appear in H_i' also lead to renormalizations of ω_0 and $V_{\kappa ss}$ (including anharmonicity); moreover, terms $b_s^+ b_s a_k^+ a_{k'}$ lead to the well-known phonon modulation broadening of the NP line $\gamma_m^{(ph)}$ (see Refs. 2-7). All these broadening mechanism lead to a total profile of the NP line which is a convolution of the distribution calculated in the present paper with a Lorentzian distribution of width $2(\gamma_d+\gamma_m^{(ph)})$. We shall also assume that ω_0 and $V_{\kappa ss}$ in Eq. (1) are renormalized accordingly.

The cross section $\sigma_{xx}(\omega)$ of the impurity absorption of light polarized parallel to the x axis is expressed in terms of the spectral representation of the time correlation function of the dipole moment operator M(t) (see Ref. 13)

$$\sigma_{xx}(\omega)=C'N\left[1-\exp\left(-\frac{\omega}{T}\right)\right]\int_{-\infty}^{\infty} dt e^{i\omega t}\langle M_x(t) M_x(0)\rangle, \quad C'=\frac{2\pi\omega}{cn(\omega)}. \quad (3)$$

Here, $k_B = 1$; ME is an operator describing the interaction of electrons at a center with the electric field of the wave; c is the velocity of light; $n(\omega)$ is the refractive index; N is the concentration of centers; $\langle\ldots\rangle$ denotes quantum statistical averaging.

Expressing M_x in terms of the electron operators $M_x = mb_1^+ b_2 + \text{H.c.}$, we can write $\sigma_{xx}(\omega)$ in the maximum region ($\omega \approx \omega_0$) in the form

$$\left.\begin{aligned} \sigma_{xx}(\omega)\approx C s(\omega), \quad C=\frac{4\pi^2\omega N m^2}{cn(\omega)}\left[1-\exp\left(-\frac{\omega}{T}\right)\right], \\ s(\omega)=\frac{1}{\pi}\,\mathrm{Re}\int_0^\infty dt e^{i\omega t} s(t), \quad s(t)=\langle b_1^+(t) b_2(t) b_2^+(0) b(0)\rangle. \end{aligned}\right\} \quad (4)$$

It is convenient to introduce in Eq. (4) the interaction representation and treat the interaction H_i'' of localized modes with phonons as a perturbation. We shall define an operator

$$\left.\begin{aligned} U(t)=\exp[i(H_0+H_i')t]\exp[-iHt]=T\exp\left[-i\int_0^t H_i''(\tau)\,d\tau\right], \\ H_i''(\tau)=\exp[i(H_0+H_i')\tau] H_i'' \exp[-i(H_0+H_i')\tau]. \end{aligned}\right\} \quad (5)$$

The correlation function $\tilde{s}(t)$ can be then written in the form

$$\left.\begin{aligned} s(t)=\exp(-i\omega_0 t) Z_e^{-1}\exp\left(-\frac{E_1}{T}\right) Q(t), \\ Q(t)=\sum_{\ldots n_\varkappa\ldots}(\ldots n_\varkappa\ldots|g(t)|\ldots n_\varkappa\ldots), \quad g(t)=\langle(2|G(t)|1)\rangle_{ph}, \\ G(t)=\frac{Z_e}{Z}\exp\left(-i\sum_\varkappa V_\varkappa a_\varkappa^+ a_\varkappa t\right) U(t) b_2^+ b_1 \exp\left(-\frac{H-E_1}{T}\right) U^+(t), \\ V_\varkappa = V_{\varkappa 22}-V_{\varkappa 11}. \end{aligned}\right\} \quad (6)$$

Here, Z is the total partition function of the system; $Z_e=\sum_i \exp(-E_i/T)$ is the electron part of the partition function which can be separated since the interaction is weak; the matrix element (2|G|1) is evaluated between the wave functions |s) of the isolated electron subsystem; $\langle\ldots\rangle_{ph}$ denotes averaging over states of the phonon thermostat; $|\ldots n_\kappa\ldots)$ is the wave function of the isolated subsystem of local vibrations.

It follows from Eqs. (4) and (6) that the profile of the NP line is determined by the spectral distribution of the function Q(t)

$$\left.\begin{aligned} Q(\Omega)=\frac{1}{\pi}\,\mathrm{Re}\int_0^\infty dt e^{i\Omega t} Q(t), \quad \Omega=\omega-\omega_0, \\ s(\omega)=Z_e^{-1}\exp\left(-\frac{E_1}{T}\right) Q(\Omega) \end{aligned}\right\} \quad (7)$$

The main contribution to the integral in Eq. (7) in the relevant region of small $|\Omega| \ll \omega_0$, ω_κ is due to large t. We can calculate the function Q(t) in this region using the method developed for caclulating the spectral distribution of nonlinear oscillators interacting with one another and with a medium.[11,12]

As in the case of nonlinear oscillators, the spectral distribution for our system of impurity electrons interacting with oscillators is a superposition of a large nubmer of partial spectra corresponding to transitions with close frequencies. Such partial spectra are not independent and the interference of various transitions is important.[12] The set of transition frequencies $\omega_0+\sum_{\varkappa'} V_{\varkappa'} n_{\varkappa'}$ corresponds to a set of occupation numbers $n_{\kappa'}$ of oscillators interacting with electrons. A similar set of frequen-

cies for a system of oscillators is determined by the interaction of a chosen oscillator κ absorbing light with the other oscillators κ', but there is also an additional frequency scatter since optical transition of the nonlinear oscillator κ can take place from different states n_κ. Only if the oscillator κ absorbing light is a high-frequency oscillator ($\omega_\kappa \gg T$), we may assume that only its lowest state $n_\kappa = 0$ is occupied and the problem of calculating its spectral distribution is equivalent to the corresponding problem for a two-level electron system interacting with localized modes. As in Ref. 11 and 12, the problem reduces to a system of differential-difference equations for the matrix elements of g(t), which is analogous to a quantum transport equation.

Following the method of Refs. 11 and 12 (see the Appendix F of Ref. 12), we can derive an operator equation for g(t). The operator equation takes the form

$$\left.\begin{aligned}\frac{\partial g}{\partial t} &= -i\sum_x V_x a_x^+ a_x g - \sum_x \{(\bar n_x+1)(a_x^+ a_x g - 2a_x g a_x^+ + g a_x^+ a_x)\\ &+ \bar n_x (a_x a_x^+ g - 2a_x^+ g a_x + g a_x a_x^+)\}\Gamma_x - i\sum_x [a_x^+ a_x,\ g]P_x,\\ \bar n_x &= \left[\exp\left(\frac{\omega_x}{T}\right)-1\right]^{-1}.\end{aligned}\right\} \quad (8)$$

Here, Γ_κ is the decay damping of localized transitions due to the interaction with phonons and P_κ is the frequency shift

$$\Gamma_x + iP_x = \int_0^\infty dt \exp(i\omega_x t - \varepsilon t)\langle [h_x(t),\ h_x(0)]\rangle_{ph}, \quad \varepsilon \to +0. \quad (9)$$

The initial condition imposed on Eq. (8) is given by

$$g(0) = \prod_x (\bar n_x + 1)^{-1} \exp\left(-\frac{\omega_x}{T} a_x^+ a_x\right). \quad (10)$$

Small terms V_κ/ω_κ, $\Gamma_\kappa/\omega_\kappa$, and $V_\kappa \partial \ln \Gamma_\kappa/\partial\omega_\kappa$ and analogous terms with P_κ have been neglected in Eqs. (8)-(10).

Passing from the operator equation (8) to an equation for the matrix elements $(\ldots n_\kappa \ldots |g(t)| \ldots n_\kappa \ldots)$ and solving the resultant system by the method of generating functions,[11, 12] we obtain the following explicit expression for Q(t):

$$\left.\begin{aligned}Q(t) &= \prod_x \exp\left(\Gamma_x t + \frac{1}{2} i V_x t\right)\psi_x^{-1}(t),\\ \psi_x(t) &= \operatorname{ch} a_x t + \left[1 + i\frac{V_x}{2\Gamma_x}(2\bar n_x + 1)\right]\frac{\Gamma_x}{a_x}\operatorname{sh} a_x t.\end{aligned}\right\} \quad (11)$$

Here,

$$a_x = \left[\Gamma_x^2 + i(2\bar n_x + 1)\Gamma_x V_x - \frac{1}{4}V_x^2\right]^{1/2}, \quad \operatorname{Re} a_x > 0 \quad (12)$$

Equation (11) is analogous to the time correlation function of a nonlinear oscillator interacting with other oscillators and with the surrounding medium.[11]

2. DISCUSSION OF THE PROFILE AND FINE STRUCTURE OF A NO-PHONON LINE

It can be seen from Eqs. (4), (6), and (11) that the spectrum of an NP line $\tilde\sigma(\omega)$ is determined by an integral of an elementary function. The spectral distribution $\tilde\sigma(\omega) \propto Q(\Omega)$ depends on the parameters on the parameters V_κ, Γ_κ, and $\bar n_\kappa$ and can be calculated numerically given any values of these parameters. Simple analytic expressions for $Q(\Omega)$ or $\tilde\sigma(\omega)$ can be obtained in some limiting cases.

When the electron–phonon interaction coupling parameter V_κ is small or the damping Γ_κ is large and the condition $|V_x|\left(\bar n_x + \frac{1}{2}\right) \ll \Gamma_x$ is satisfied, we can expand Eqs. (12) and (11) in powers of V_κ/Γ_κ up to fourth-order terms in V_κ/Γ_κ. Substituting such expansions in Eq. (7) we obtain

$$\left.\begin{aligned}Q(\Omega) &= Q'(\Omega) - Q''(\Omega),\\ Q'(\Omega) &= \frac{1+\varepsilon_1}{\pi}\frac{\gamma' + \varepsilon_2\Omega'}{\gamma'^2 + \Omega'^2}, \quad Q''(\Omega) = \sum_x \varepsilon_x Q_x''(\Omega),\\ Q_x''(\Omega) &= \frac{1}{\pi}\frac{\gamma_x + \varepsilon_x'\Omega_x}{\gamma_x^2 + \Omega_x^2}, \quad |V_x|\left(\bar n_x + \frac{1}{2}\right) \ll \Gamma_x,\end{aligned}\right\} \quad (13)$$

$$\begin{aligned}&\Omega' = \Omega - \sum_x (\bar n_x V_x - \varepsilon_x'\Gamma_x), \quad \gamma' = 2\sum_x \varepsilon_x \Gamma_x\left[1 - \frac{V_x^2}{4\Gamma_x^2}(5\bar n_x^2 + 5\bar n_x + 1)\right],\\ &\Omega_x = \Omega - V_x - \sum_{x_1} V_{x_1}\bar n_{x_1}(1 + 2\delta_{xx_1}), \quad \gamma_x = 2\Gamma_x + 2\sum_{x_1}\varepsilon_{x_1}\Gamma_{x_1}(1 + 2\delta_{xx_1}),\\ &\varepsilon_1 = \sum_x \varepsilon_x, \quad \varepsilon_2 = \sum_x \varepsilon_x\varepsilon_x', \quad \varepsilon_x = \frac{1}{4}\frac{V_x^2}{\Gamma_x^2}\bar n_x(\bar n_x + 1), \quad \varepsilon_x' = \frac{V_x}{\Gamma_x}(2\bar n_x + 1).\end{aligned} \quad (14)$$

The parameters ε in Eq. (13) are now small quantities. The term $Q'(\Omega)$ near the maximum of the spectral distribution resembles a Lorentzian function of width $2\gamma'$ at half the height and with a maximum at $\Omega_m \approx \sum_x \bar n_x V_x$. Retaining in the expressions for γ', Ω' only the first nonvanishing terms in V_κ/Γ_κ, we obtain $Q'(\Omega)$ which reduces to the result of Refs. 4 and 5. The width of the peak $2\gamma'$ is determined by V_κ^2/Γ_κ and is small compared with the damping of localized modes Γ_κ. The function $Q''(\Omega)$ in Eq. (13) has a considerable width $\sim\Gamma_\kappa \gg \gamma'$ and is small in the central region of the peak compared with $Q'(\Omega)$. However, in the wings of the distribution ($|\Omega| \gg \Gamma_\kappa$) $Q''(\Omega)$ we find that $Q''(\Omega)$ in comparable with $Q'(\Omega)$ and these two terms in $Q(\Omega)$ largely cancel one another.

We shall now discuss the opposite limiting case $|V_\kappa| \gg 2\Gamma_\kappa(2\bar n_\kappa + 1)$. It is convenient to transform Eq. (11) for Q(t) by separating in $\psi_\kappa(t)$ a factor $\exp(a_\kappa t)$ and expanding $\psi_\kappa^{-1}(t)\exp(a_\kappa t)$ in powers

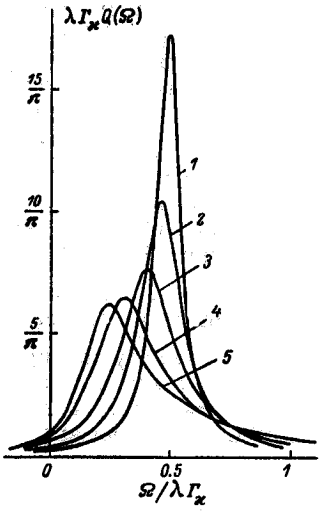

FIG. 1. Spectral distributions $Q(\Omega)$ of an impurity electron interacting with a local vibration κ in the classical limit: curves 1-5 correspond to the values of λ = 0.5, 1, 2, 5, and 10.

of $\exp(-2a_\kappa t)$. Integrating with respect to t in Eq. (7), we arrive at the following exact expression for $Q(\Omega)$ in the form of a series:

$$Q(\Omega)=\frac{1}{\pi}\sum_{\mathbf{m}}|D(\mathbf{m})|\frac{\Gamma(\mathbf{m})\cos\alpha(\mathbf{m})-\Omega(\mathbf{m})\sin\alpha(\mathbf{m})}{\Gamma^2(\mathbf{m})+\Omega^2(\mathbf{m})}, \qquad (15)$$

$$D(\mathbf{m})=\prod_x\frac{16a_x\Gamma_x}{4(a_x+\Gamma_x)^2+V_x^2}\left[\frac{4(a_x-\Gamma_x)^2+V_x^2}{4(a_x+\Gamma_x)^2+V_x^2}\right]^{m_x},\quad \alpha(\mathbf{m})=ArgD(\mathbf{m}),$$

$$\Omega(\mathbf{m})=\Omega+\frac{1}{2}\sum_x V_x-\sum_x(2m_x+1)a_x'',$$

$$\Gamma(\mathbf{m})=\sum_x[(2m_x+1)a_x'-\Gamma_x],\quad a_x\equiv a_x'+ia_x'',\quad \mathbf{m}=(\ldots m_x\ldots). \qquad (16)$$

The summation over m in Eq. (15) is over all values of $m_\kappa = 0, 1, 2, \ldots$.

Equation (15) is the resultant spectral distribution representing a set of partial spectra corresponding to transitions in the electron subsystem for different occupation numbers of localized modes m_κ. Since the frequencies of such transitions are close to one another, the transitions interfere and, therefore, the amplitudes and profiles of partial spectra may differ considerably from the properties of the spectra in systems with very different transition frequencies.[12]

In the limit of small Γ_κ or large $|V_\kappa|$, the interference is largely suppressed and various parameters in Eqs. (15) and (16) simplify

$$\left.\begin{aligned}
&|D(\mathbf{m})|=\prod_x(\bar n_x+1)^{-1}\exp\left(-\frac{\omega_x m_x}{T}\right),\\
&\alpha(\mathbf{m})=4\sum_x\frac{\Gamma_x}{V_x}(m_x-\bar n_x),\\
&\Omega(\mathbf{m})=\Omega-\sum_x V_x m_x-4\sum_x(2m_x+1)\bar n_x(\bar n_x+1)\frac{\Gamma_x^2}{V_x},\\
&\Gamma(\mathbf{m})=\sum_x\left[(2m_x+1)(2\bar n_x+1)\left(1-8\bar n_x(\bar n_x+1)\frac{\Gamma_x^2}{V_x^2}\right)-1\right]\Gamma_x,\\
&|V_x|\gg 2\Gamma_x(2\bar n_x+1).
\end{aligned}\right\} \qquad (17)$$

The partial spectra then overlap only weakly and the spectral distribution exhibits a fine structure. It consists of a set (with respect to κ) of lines corresponding to different m_κ and separated from one another by $\approx|V_\kappa|$. The line widths are determined by Γ_κ for all excited vibrations and are proportional to m_κ and $\bar n_\kappa$. The asymmetry of the lines is determined by the parameters $\alpha(\mathbf{m})$ and is small.

In addition to the limiting cases of large and small values of the ratio $\Gamma_\kappa/|V_\kappa|$, we can also consider the limiting cases of low and high temperatures. The condition $\bar n_\kappa \ll 1$ is satisfied at low temperatures and the term with $m_\kappa = 0$ is most important in the expansion (15); the terms with $m_\kappa = 1$, and $m_{\kappa'} = 0$ ($\kappa' \neq \kappa$) in the region of a maximum of the distribution lead only to small corrections. The resultant distribution is given by Eq. (13), where we can now neglect terms $\sim\bar n_\kappa$ compared with unity

$$\left.\begin{aligned}
&\Omega'=\Omega-4\sum_x V_x\mu_x\bar n_x,\quad \gamma'=2\sum_x v_x^2\mu_x\Gamma_x\bar n_x,\\
&\Omega_x=\Omega-V_x,\quad \gamma_x=2\Gamma_x,\quad \varepsilon_1=\sum_x\varepsilon_x,\quad \varepsilon_2=4\sum_x v_x^2\mu_x^2\bar n_x,\\
&\varepsilon_x=v_x^2\mu_x^2(4-v_x^2)\bar n_x,\quad \varepsilon_x'=\frac{4v_x}{4-v_x^2},\quad v_x=\frac{V_x}{\Gamma_x},\quad \mu_x=\frac{1}{4+v_x^2},\quad \bar n_x\ll 1.
\end{aligned}\right\} \qquad (18)$$

A fine structure appears in the distribution defined by Eqs. (13) and (18) for $|V_\kappa| \gg 2\Gamma_\kappa$. Almost all the intensity is in the peak $Q'(\Omega)$ and in the peaks at frequencies shifted by $|V_\kappa|$ from the principal peak, i.e., the peaks $\varepsilon_\kappa Q_\kappa''(\Omega)$ have low total intensities (proportional to $\bar n_\kappa$). The widths of such weak peaks $4\Gamma_\kappa$ are determined by damping of localized modes and the width of the strong peak $2\gamma' \approx 4\sum_x\Gamma_x\bar n_x$ for small $\bar n_\kappa$ is much smaller and, therefore, this peak is much higher than the weak peaks (by a factor of $\bar n_\kappa^{-2}$ provided there is only one localized mode). It follows from Eqs. (18) or (17) that the intensities of weak peaks are comparable with the intensities of the wings of the narrow peak with $m_\kappa = 0$. The structure of the spectrum becomes smeared with decreasing $|V_\kappa|/\Gamma_\kappa$ and disappears eventually (for $|V_\kappa| \sim \Gamma_\kappa$).

The condition of existence of a fine structure (17) ceases to be satisfied at high temperatures and the distribution $Q(\Omega)$ becomes smooth. We shall now discuss the distribution $Q(\Omega)$ in the classical limit $\hbar \to 0$ when the conditions

$$|V_x|\ll\Gamma_x,\quad \bar n_x\approx T/\hbar\omega_x\gg 1\ (V_x\sim\hbar),$$

are satisfied and restrict ourselves for simplicity to the case of interaction with a single localized mode κ. It follows from Eqs. (7), (11) and (12) that the spectral distribution $Q(\Omega)$ regarded as a function of the dimensionless variable Ω/Γ_κ depends on a single dimensionless parameter

$$\lambda=\frac{V_x(2\bar n_x+1)}{\Gamma_x}\approx\frac{2V_xT}{\Gamma_x\omega_x}. \qquad (19)$$

The distribution $Q(\Omega)$ can be studied analytically in the limiting cases $|\lambda| \ll 1$ and $|\lambda| \gg 1$. In the first case, $Q(\Omega)$ is determined by Eqs. (13) and (14) where all the terms $\sim V_\kappa$ together with $V_\kappa\bar n_\kappa \approx V_\kappa T/\omega_\kappa$ should be neglected. For $|\lambda| \gg 1$, the spectral distribution in the region $\Omega/\lambda \gtrsim \Gamma_\kappa$ is formed during a time $t \lesssim (\Gamma_\kappa|\lambda|)^{-1} \ll |a_\kappa|^{-1} \sim (\Gamma_\kappa\sqrt{|\lambda|})^{-1}$. We then have $\psi_x(t)\approx 1+\frac{1}{2}i\Gamma_x\lambda t$. in Eq. (11). Integrating with respect to t in Eq. (7), we obtain

$$Q(\Omega)=\frac{2}{\Gamma_x|\lambda|}\exp\left(-\frac{2\Omega}{\Gamma_x\lambda}\right),\quad \frac{\Omega}{\lambda}\gtrsim\Gamma_x,\quad |\lambda|\gg 1,\ \bar n_x\gg 1. \qquad (20)$$

Equation (20) is valid for large $\bar n_\kappa$ and for $|V_\kappa| \gtrsim \Gamma_\kappa$. The distribution defined by Eq. (20)

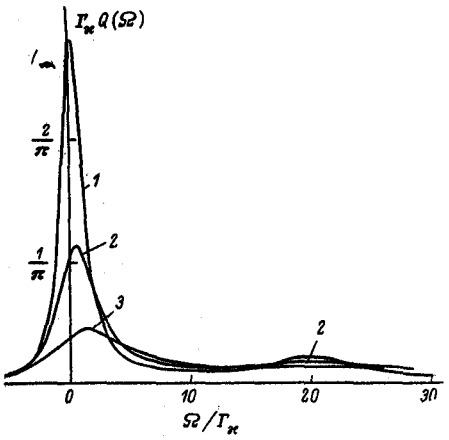

FIG. 2. Spectra of a no-phonon line corresponding to interaction with a single localized vibration κ for $V_\kappa/\Gamma_\kappa = 20$; curves 1-3 correspond to occupation numbers $\bar n_\kappa = 0.5$, 1, and 2 ($k_BT/\hbar\omega_\kappa \approx 0.91$, 1.44, 2.47).

is the envelope of a set of lines of fine structure defined by Eq. (17). Long times make an important contribution to $Q(\Omega)$ in the region $\Omega/\lambda \ll \Gamma_K$ and Eq. (20) is no longer applicable. It can be shown that the spectral distribution reaches a maximum of height $\approx 2/\Gamma_K|\lambda|$ in this region and then decreases rapidly with decreasing Ω/λ. It follows that the distribution $Q(\Omega)$ is highly asymmetric. It can be seen from Eq. (20) that the characteristic width of the distribution $\sim|V_K|\bar{n}_K$ is independent of the damping Γ_K.

The form of the spectral distribution in the region of its wings can be easily discussed for arbitrary values of the parameters and temperatures and it is also possible to find the first two moments of the function $Q(\Omega)$. These properties of the spectrum are determined by the behavior of the function $Q(t)$ during short times. Expanding Eq. (11) in powers of t and using Eq. (7), we find that $Q(\Omega)$ decreases in the wings as Ω^{-4}

$$Q(\Omega)=\frac{2}{\pi}\sum_x \Gamma_x V_x^2 \bar{n}_x(\bar{n}_x+1)\frac{1}{\Omega^4}\quad (|\Omega|\gg\Gamma_x,\ |V_x|(2\bar{n}_x+1)). \tag{21}$$

The first and second centered moments M_1 and M_2 of the function $Q(\Omega)$ are given by

$$M_1=\sum_x V_x\bar{n}_x,\quad M_2=\sum_x V_x^2\bar{n}_x(\bar{n}_x+1). \tag{22}$$

It follows that M_2 is independent of Γ_K, i.e., of the interaction of localized modes with the medium. For $|V_x|\left(\bar{n}_x+\frac{1}{2}\right)\ll\Gamma_x$ it can be seen from Eqs. (13) and (14) that the distribution $Q(\Omega)$ has a narrow peak of width $2\gamma' \ll \sqrt{M_2}$ and the second moment of the spectrum is formed due mainly to wide smooth wings of the spectrum. The characteristic width of the central region of the distribution for $|V_K| \gg \Gamma_K$ is given by $\sim\sqrt{M_2}$. Assuming that electrons interact strongly with a large number $\nu \gg 1$ of localized modes, we find that the distribution $Q(\Omega)$ for $|V_K| \gg \Gamma_K$ is described in the central region by a Gaussian function with a width at half its height $4\sqrt{\nu \ln 2 M_2}$.

The spectral distributions for intermediate values of the parameters, where simple approximate expressions cannot be obtained, may be studied numerically. It is most convenient to base any numerical study on Eq. (15). Our numerical calculations were made assuming interaction with a single localized vibration.

The results of our calculation of $Q(\Omega)$ in the classical limit $|V_K| \ll \Gamma_K$, $\bar{n}_K \gg 1$ for a range of parameters λ in Eq. (19) are shown in Fig. 1. It can be seen from Fig. 1 that the width of the distribution increases with increasing λ (in units of $2|V_K|\bar{n}_K$) and its asymmetry and maximum are shifted toward smaller Ω/V_K. Curve 1 for $\lambda = 0.5$ agrees with the analytic results (13) and (14) and curve 5 in its smooth right wing is described by Eq. (20), but it also shows the behavior of $Q(\Omega)$ in its maximum region to the left of the wing.

To illustrate the fine structure of an NP line and its smearing with increasing temperature, we show in Fig. 2 $Q(\Omega)$ calculated for a large value $V_K/\Gamma_K = 20$. It can be seen that a second peak is weak at all temperatures considered. It appears in a range of temperatures bounded from above and below.

It follows from our numerical calculations that the dependences of the shift of the peak maximum on the temperature and ratio $|V_K|/\Gamma_K$ are weaker in the region $\bar{n}_K \sim 1$ than the corresponding dependences of its width. The temperature dependences of the positions and widths of peaks for a similar problem of the spectra of localized vibrations of adsorbed molecules were calculated in Ref. 14 for a range of parameters characterizing such a system.

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