

*TEMPERATURE DEPENDENCE OF THE ABSORPTION LINE WIDTH OF LOCAL OSCILLATIONS OF H<sup>-</sup> AND D<sup>-</sup> IONS IN ALKALI HALIDE CRYSTALS*

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Infrared absorption by local oscillations in H<sup>-</sup> and D<sup>-</sup> ions in alkali halide crystals is considered. It is shown that the largest contribution to the line width is from scattering of the oscillation of the fundamental lattice by impurity atoms. The conclusions regarding the temperature dependence of the line width are in satisfactory agreement with the experimental data now available.

THE introduction of even a small number of impurities into a crystal changes radically the spectrum of its lattice vibrations. In particular, for light impurities, with mass  $M'$  much smaller than the mass  $M$  of the atom of the host lattice, local vibrations occur in the lattice spectrum, with frequency  $\omega_{loc}$  larger than the maximum frequency of the pure lattice  $\omega_L$ <sup>[1-3]</sup>. These oscillations produce in the crystal a first-order electric moment, which leads to resonant absorption of light at the frequency  $\omega_{loc}$ .

Such absorption was first observed experimentally by Schaefer<sup>[4]</sup>, who introduced H<sup>-</sup> and D<sup>-</sup> ions in alkali-halide crystals. A detailed study of the temperature dependence of the line width of this resonance absorption was carried out by Mirlin and Reshina<sup>[5]</sup> for KCl and KBr and by Hayes et al.<sup>[6]</sup> for CaF<sub>2</sub>. They found that the temperature dependence of the line width is intermediate between linear and quadratic, that is, it is of the same type as the temperature dependence of the line width of the fundamental absorption by the pure lattice<sup>[7]</sup>. It can therefore be assumed that in this case, too, the broadening can be attributed to anharmonicity of the lattice vibrations.

The H<sup>-</sup> ion produced in the investigated case is a local level with frequency  $\omega_H > 2\omega_L$ . It is obvious that the width of this level  $\gamma_H$  can be determined only by the fourth-order anharmonic term. The D<sup>-</sup> ion gives a local frequency  $\omega_D < 2\omega_L$ . The width of this level  $\gamma_D$  can be governed by anharmonic terms of either third or fourth order. It would be natural here to expect that  $\gamma_H < \gamma_D$ . Experiment shows, however, that this relation is true only for temperatures  $T$  much lower than the Debye temperature  $\Theta$ . At high tempera-

tures the width  $\gamma_H$  turns out to be larger than  $\gamma_D$ . Another curious fact is that the line widths depend in marked fashion on the temperature when  $T < \Theta$ .

A theoretical analysis of the local-peak width connected with third-order anharmonic terms was carried out by a number of authors<sup>[8-13]</sup>. The influence of fourth-order anharmonicity, which leads to decay of local oscillations into three phonons of the host lattice, was investigated by Zavt et al.<sup>[14]</sup> Hayes et al.<sup>[6]</sup> were first to note that due allowance for the anharmonic fourth-order terms leads to processes corresponding to anharmonic interaction between the local oscillations themselves. There is a process that describes "scattering" of the oscillation of the host lattice by the impurity atom. The corresponding energy conservation law is of the form  $\omega_{loc} + \omega_1 = \omega_{loc} + \omega_2$ . It was indicated in<sup>[6]</sup> that observed experimental relations can be attributed to a process of this type. Recently Krivoglaz et al.<sup>[15]</sup> obtained an explicit expression for the probability of such a process. A general theory of the width of the local level, developed by Maradudin<sup>[16]</sup>, contains in principle all the processes connected with allowance for the anharmonic terms of third and fourth order.

In none of the foregoing investigations, however, were attempts made to show that in alkali-halide crystals the process considered in<sup>[6]</sup> makes the greatest contribution to the width of the local level at high temperatures. The present paper is devoted to establishment of the fact that this process is decisive for the width of the local levels of H<sup>-</sup> and D<sup>-</sup> ions in alkali-halide crystals. We use a coordinate representation, in which one can readily see at each stage all the physical parameters that permit various simplifications in

the theory. The developed theory confirms qualitatively the experimentally observed regularities. Namely, at high temperatures the relative widths satisfy the relation  $\gamma_H/\gamma_D \approx (M_D/M_H)^{1/2} > 1$  for the isotopic defect considered in nearest-neighbor approximation. At low temperatures, the aforementioned "scattering" process is insignificant, and then the width ratio is reversed:  $\gamma_H/\gamma_D < 1$ . Under certain assumptions it is also possible to attribute to this scattering the dependence of the line width on the temperature when  $T < \Theta$ .

**1. ABSORPTION COEFFICIENT IN THE ANHARMONIC APPROXIMATION**

For small impurity concentrations we can consider absorption by a single impurity atom located at the origin, and multiply the result by the number of impurities in the volume under consideration. In this case the coefficient of absorption of infrared light at frequencies  $\omega \approx \omega_{loc} > \omega_L$  can be represented in the form<sup>[10,17,18]</sup>

$$K(\omega) = -c_d \frac{e^2}{\hbar v_0} \text{Im } D_{\alpha\beta}^R(0, 0; \omega) e_\alpha e_\beta, \tag{1}$$

where

$$D_{\alpha\beta}^R(l\kappa, l'\kappa'; \omega) = \int_0^\infty dt e^{i\omega t} \langle [u_\alpha(l\kappa; t), u_\beta(l'\kappa'; 0)] \rangle \tag{2}$$

is the single-particle retarded Green's function of the lattice containing the impurities. Here  $u_\alpha(l\kappa; t)$  is the  $\alpha$ -component of the displacement of the  $\kappa$ -th atom in the  $l$ -th unit cell at the instant of time  $t$ ; the angle brackets denote the averaging  $\langle \dots \rangle = \text{Sp}(e^{-\beta H} \dots)$ ;  $c_d$  is the impurity concentration,  $e$  the effective charge of the impurity atom,  $b_0$  the unit-cell volume, and  $e_\alpha$  the unit vector of light polarization.

An essential factor in the derivation of (1), as shown by Kagan and Iosilevskii<sup>[9]</sup>, is that when a light impurity with mass  $M'$  much smaller than the mass of the host-lattice atom  $M$  is introduced, the maximum amplitude at the frequency  $\omega_{loc}$  is possessed by the oscillation of the impurity atom, whereas the displacements of the atoms of the host lattice are small. This result was also obtained by Montroll and Potts<sup>[20]</sup> for a one dimensional model of the lattice and by Maradudin<sup>[21]</sup> for a single-atom lattice. The latter has shown that terms of the type  $D_{\alpha\beta}^R(0, l; \omega)$ , which depend on the host-lattice atom positions,  $l \neq 0$ , contain small exponential factors of the type

$$\exp\{-[2(\omega_{loc}^2 - \omega_L^2)/\Delta^2]^{1/2} R_{0l}/a\},$$

where  $\Delta$  is the dispersion of the  $\omega_L$  branch,  $R_{0l}$  is the distance from the impurity atom to the  $l$ -th

atom, and  $a$  is the lattice constant. This smallness will be all the more pronounced the lighter the mass of the impurity atom. Obviously, this approximation is more suitable for  $H^-$  as an impurity ion than for  $D^-$ .

The Green's function  $D_{\alpha\beta}^R(0, 0; \omega)$  in (1) differs from the Green's function of an ideal lattice  $D_{\alpha\beta}^{(0)R}(l\kappa, l'\kappa'; \omega) = \frac{\hbar}{(M_\kappa M_{\kappa'})^{1/2} N} \sum_{\mathbf{k}_j} \frac{\xi_\alpha(\kappa|\mathbf{k}_j) \xi_\beta^*(\kappa'|\mathbf{k}_j)}{\omega^2 - \omega_{\mathbf{k}_j}^2 + i\delta \text{sign } \omega} \times \exp\{i\mathbf{k}(\mathbf{R}_l - \mathbf{R}_{l'})\}$  (3)

even in the harmonic approximation, owing to the mass difference between the impurity and host atoms, and owing to the change in the force constants. In (3)  $M_\kappa$  is the mass of the host-lattice atom located at site  $\kappa$ ,  $\xi_\alpha(\kappa|\mathbf{k}_j)$  is the  $\alpha$ -component of the polarization vector of the phonon of branch  $j$  with wave vector  $\mathbf{k}$ , and  $N$  is the number of unit cells in the crystal volume under consideration.

For the mass defect in a cubic crystal, the function  $D_{\alpha\beta}^R = D^R \delta_{\beta\alpha}$  is equal to<sup>[10,21]</sup>

$$D_{\alpha\beta}^R(0, 0; \omega) = \frac{D^{(0)R}(0, 0; \omega) \delta_{\alpha\beta}}{1 - \delta L(0, 0; \omega) D^{(0)R}(0, 0; \omega)},$$

$$\delta L(0, 0; \omega) \delta_{\alpha\beta} = \frac{M - M'}{\hbar} \omega^2 \delta_{\alpha\beta} = \frac{M}{\hbar} \varepsilon \omega^2 \delta_{\alpha\beta}. \tag{4}$$

It follows from (1) and (4) that  $K(\omega)$  has a  $\delta$ -type peak at frequency  $\omega_{loc}$ , with  $\omega_{loc}$  the solution of the equation

$$1 = \delta L(0, 0; \omega) D^{(0)R}(0, 0; \omega). \tag{5}$$

Allowance for the anharmonic terms in the Hamiltonian of the lattice leads, naturally, to the appearance of a finite width of this peak and to a shift of the resonant frequency. The problem of calculating the finite line width at the frequency  $\omega \approx \omega_{loc}$  reduces to a calculation of the Green's function of the lattice  $\tilde{D}_{\alpha\beta}^R(0, 0; \omega)$  with allowance for the anharmonic terms of third and fourth orders. It is convenient to use for the calculations the method of temperature Green's functions, for which a diagram technique exists<sup>[22]</sup>. The temperature Green's function  $\tilde{D}_{\alpha\beta}(0, 0; i\omega_n)$  (where  $\omega_n = 2\pi nT/\hbar$ ) satisfies the Dyson equation:

$$\tilde{D}_{\alpha\beta}(0, 0; i\omega_n) = D_{\alpha\beta}(0, 0; i\omega_n) + D_{\alpha\gamma}(0, l\kappa; i\omega_n) \Pi_{\gamma\delta}(l\kappa, l'\kappa'; i\omega_n) \tilde{D}_{\delta\beta}(l'\kappa', 0; i\omega_n); \tag{6}$$

here  $\Pi_{\gamma\delta}(l\kappa, l'\kappa'; i\omega_n)$  is the polarization operator connected with the presence of the anharmonic terms; summation over all the repeated indices is implied. This equation differs from that obtained by Maradudin<sup>[16]</sup> in that it is written in the coordinate representation.

The function  $\tilde{D}_{\alpha\beta}^R(l\kappa, l'\kappa'; \omega)$  is an analytic continuation, in terms of  $\omega$ , of the temperature Green's function  $\tilde{D}_{\alpha\beta}(l\kappa, l'\kappa'; i\omega_n)$ . Using the same small parameter as in the derivation of (1), we neglect in the expression for  $\tilde{D}^R(0, 0; \omega_{10c})$  the terms with  $D_{\alpha\beta}^R(l\kappa, 0; \omega_{10c})$  and  $D_{\alpha\beta}^R(l\kappa, l'\kappa'; \omega_{10c})$ , compared with the terms containing  $D_{\alpha\beta}^R(0, 0; \omega_{10c})$ . Taking also into account the fact that for crystals with cubic symmetry

$$\begin{aligned} D_{\alpha\beta}^R(0, 0; \omega) &= D^R(0, 0; \omega) \delta_{\alpha\beta}, \\ \Pi_{\alpha\beta}(0, 0; \omega) &= \Pi_{\alpha\beta}(0, 0; \omega) \delta_{\alpha\beta}, \end{aligned} \quad (7)$$

we obtain

$$\tilde{D}^R(0, 0; \omega) = \frac{D^R(0, 0; \omega)}{1 - D^R(0, 0; \omega) \Pi(0, 0; \omega)}. \quad (8)$$

Or, introducing  $\Pi = \Delta - i\Gamma$  and using (4), we get ultimately

$$\begin{aligned} \tilde{D}^R(0, 0; \omega) &= D^{(0)R}(0, 0; \omega) \{1 - D^{(0)R}(0, 0; \omega) [\delta L(0, 0; \omega) \\ &+ \Delta(0, 0; \omega)] + iL^{(0)R}(0, 0; \omega) \Gamma(0, 0; \omega)\}^{-1}. \end{aligned} \quad (9)$$

The new shifted position  $\omega_{10c} = \omega_{10c} + \delta\omega$  should be obtained from the equation

$$1 - D^{(0)R}(0, 0; \omega) [\delta L(0, 0; \omega) + \Delta(0, 0; \omega)] = 0. \quad (10)$$

From (9) and (4) we see that the quantity  $D^{(0)R}(0, 0; \omega) \Gamma(0, 0; \omega) / 2(1 - \epsilon)$  plays the role of the relative width of the local peak  $\gamma(\omega_{10c})$ . Using (1), (9) and (10), we obtain an expression for the absorption coefficient at the maximum:

$$K(\omega_{10c}) = c_d \frac{e^2}{\hbar\nu_0} \frac{1}{\Gamma(0, 0; \omega_{10c})}. \quad (11)$$

### 3. CALCULATION OF $\Gamma(0, 0; \omega)$ AND DISCUSSION OF RESULTS

For the calculation of  $\Gamma(0, 0; \omega) = -\text{Im} \Pi(0, 0; \omega)$  it is convenient, just as in [23], to express  $D_{\alpha\beta}(l\kappa, l'\kappa'; i\omega_n)$  in terms of the spectral density  $\rho_{\alpha\beta}(l\kappa, l'\kappa'; \omega)$ :

$$\begin{aligned} D_{\alpha\beta}(l\kappa, l'\kappa'; i\omega_n) &= \int_{-\infty}^{\infty} \frac{d\omega'}{\pi} \rho_{\alpha\beta}(l\kappa, l'\kappa'; \omega') \\ &\times \left[ \frac{1}{i\omega_n - \omega'} - \frac{1}{i\omega_n + \omega'} \right], \end{aligned} \quad (12)$$

$$\rho_{\alpha\beta}(l\kappa, l'\kappa'; \omega) = -\text{Im} D_{\alpha\beta}^R(l\kappa, l'\kappa'; \omega). \quad (13)$$

The contribution to the width of the local peak is made by diagrams of the same type as in the case of a pure lattice [7]. Now, however, the lines correspond to the Green's function of the impurity lat-

tice. Calculations in accordance with the usual rules [22] yield

$$\begin{aligned} \Gamma_{\alpha\alpha}^{(3)}(0, 0; \omega_{10c}) &= 3^2(3-1)! \sum \Phi_{\alpha\beta\gamma}(0, l_1\kappa_1, l_2\kappa_2) \\ &\times \Phi_{\alpha\delta\rho}(0, l_1'\kappa_1', l_2'\kappa_2') \\ &\times \int_0^{\infty} \frac{d\omega_1 d\omega_2}{\pi^2} \rho_{\beta\delta}(l_1\kappa_1, l_1'\kappa_1'; \omega_1) \rho_{\gamma\rho}(l_2\kappa_2, l_2'\kappa_2'; \omega_2) \\ &\times \{\delta(\omega_{10c} - \omega_1 - \omega_2)[1 + N_1 + N_2] \\ &+ 2\delta(\omega_{10c} - \omega_1 + \omega_2)[N_2 - N_1]\}, \end{aligned} \quad (14)$$

$$\begin{aligned} \Gamma_{\alpha\alpha}^{(4)}(0, 0; \omega_{10c}) &= 4^2(4-1)! \sum \Phi_{\alpha\beta\gamma\mu}(0, l_1\kappa_1, l_2\kappa_2, l_3\kappa_3) \\ &\times \Phi_{\alpha\delta\tau\nu}(0, l_1'\kappa_1', l_2'\kappa_2', l_3'\kappa_3') \\ &\times \int_0^{\infty} \frac{d\omega_1 d\omega_2 d\omega_3}{\pi^3} \rho_{\beta\delta}(l_1\kappa_1, l_1'\kappa_1'; \omega_1) \\ &\times \rho_{\gamma\tau}(l_2\kappa_2, l_2'\kappa_2'; \omega_2) \rho_{\mu\nu}(l_3\kappa_3, l_3'\kappa_3'; \omega_3) \\ &\times \{\delta(\omega_{10c} - \omega_1 - \omega_2 - \omega_3) \\ &[(1 + N_1)(1 + N_2)(1 + N_3) - N_1 N_2 N_3] \\ &+ 3\delta(\omega_{10c} + \omega_1 + \omega_2 - \omega_3)[N_1 N_2(1 + N_3) \\ &- (1 + N_1)(1 + N_2)N_3] + \\ &3\delta(\omega_{10c} + \omega_1 - \omega_2 - \omega_3)[N_1(1 + N_2)(1 + N_3) \\ &- (1 + N_1)N_2 N_3]\}, \end{aligned} \quad (15)$$

where

$$N_i = 1 / (\exp\{-\hbar\omega_i / T\} - 1).$$

Summation over  $l_1\kappa_1, l_2\kappa_2, l_3\kappa_3$ , etc. is carried out here over all lattice sites, including the position of the impurity atom  $l\kappa = 0$ . The numerical factors are of combinatorial origin; for anharmonicity of  $n$ -th order the corresponding factor is  $n^2(n-1)!$ .

Expressions (14) and (15) can be noticeably simplified in each concrete case by taking into account the fact that for light impurities the local frequencies correspond to larger impurity-atom displacement amplitudes exceeding the total amplitude of the atom oscillations at the frequencies of the continuous spectrum [12, 18, 19]. Therefore the maximal terms in (14) and (15) will be those containing  $\rho_{\alpha\beta}(0, 0; \omega_{10c})$  and  $\rho_{\alpha\beta}(l\kappa, l'\kappa'; \omega_i)$ , where  $\omega_i \leq \omega_L$  and  $l\kappa, l'\kappa' \neq 0$ .

Let us consider the concrete case of the ions  $H^-$  and  $D^-$  in alkali-halide crystals. Since for  $H^-$  we have  $\omega_{10c} > 2\omega_L$ , the width of the local peak is determined only by  $\Gamma^{(4)}$ . It is obvious here that the conservation laws for the typical spectrum of

an alkali-halide crystal admit of two processes for  $\gamma^{(4)}$ : the decay of the local oscillations into oscillations of the continuous spectrum ( $\omega_{10c} = \omega_1 + \omega_2 + \omega_3$ ), and "scattering" of the oscillations of the continuous spectrum by the impurity atom ( $\omega_{10c} + \omega_1 = \omega_{10c} + \omega_2$ ). Here  $\omega_i$  are frequencies of the continuous spectrum. Picking out the terms of (18) that are the largest in this sense, we should retain for the decay process only summations over the nonvanishing values of  $l_1\kappa_1, l_2\kappa_2, l_3\kappa_3$ , etc. For the "scattering" process it is necessary to take one of the  $l_1\kappa_i$  and  $l'_1\kappa'_i$  equal to zero, and two others not equal to zero. Then at temperatures  $T < \hbar\omega_{10c}$  we have

$$\begin{aligned} \Gamma_{\alpha\alpha}^{(4)} = & 4^2(4-1)! \sum \Phi_{\alpha\beta\gamma\mu}(0, l_1\kappa_1, l_2\kappa_2, l_3\kappa_3) \\ & \times \Phi_{\alpha\delta\tau\nu}(0, l'_1\kappa'_1, l'_2\kappa'_2, l'_3\kappa'_3) \\ & \times \int_0^\infty \frac{d\omega_1 d\omega_2 d\omega_3}{\pi^3} \rho_{\beta\delta}(l_1\kappa_1, l'_1\kappa'_1; \omega_1) \rho_{\gamma\tau}(l_2\kappa_2, l'_2\kappa'_2; \omega_2) \\ & \times \rho_{\mu\nu}(l_3\kappa_3, l'_3\kappa'_3; \omega_3) \delta(\omega_{10c} - \omega_1 - \omega_2 - \omega_3) \\ & \times [(1 + N_1)(1 + N_2)(1 + N_3) - N_1 N_2 N_3] + 4^2(4-2)! \\ & \times \sum \Phi_{\alpha\beta\gamma\mu}(0, l_1\kappa_1, l_2\kappa_2, 0) \Phi_{\alpha\delta\tau\mu}(0, l'_1\kappa'_1, l'_2\kappa'_2, 0) \\ & \times 3 \int_0^\infty \frac{d\omega_1 d\omega_2 d\omega_3}{\pi^3} \rho_{\beta\delta}(l_1\kappa_1, l'_1\kappa'_1; \omega_1) \rho_{\gamma\tau}(l_2\kappa_2, l'_2\kappa'_2; \omega_2) \\ & \times \rho_{\mu\mu}(0, 0; \omega_3) \delta(\omega_{10c} + \omega_1 - \omega_2 - \omega_3) [N_1(1 + N_2)]. \end{aligned} \quad (16)$$

In accordance with the definition (13), when  $l\kappa, l'\kappa' \neq 0$  the spectral density  $\rho_{\alpha\beta}(l\kappa, l'\kappa'; \omega)$  contains amplitudes of the displacements of the host-lattice atoms  $(\hbar/2M_K\omega)^{1/2}$  and  $(\hbar/2M_{K'}\omega)^{1/2}$ . The spectral density  $\rho_{\alpha\alpha}(0, 0; \omega)$ , which differs essentially from zero only when  $\omega = \omega_{10c}$ , contains the square of the amplitude of the impurity atom  $\hbar/2M'\omega_{10c}$ . For an order-of-magnitude estimate we can assume that summation over  $\kappa$  includes only the nearest neighbors. Then the ratio of the second term in (16) to the first is

$$\alpha \approx \frac{M_\kappa}{M'} \frac{\omega_L}{\omega_{10c}}. \quad (17)$$

Since  $\omega_{10c} \approx (M/M')^{1/2} \langle \omega^2 \rangle^{1/2}$  [2], the final estimate will be

$$\alpha \approx \left( \frac{M_\kappa^2}{M'M} \frac{\omega_L^2}{\langle \omega^2 \rangle} \right)^{1/2} > 1. \quad (18)$$

For a KCl crystal containing  $H^-$  in place of  $Cl^-$  we have  $\alpha \approx 10$ ; for KBr containing  $H^-$  in lieu of  $Br^-$ ,  $\alpha \approx 7$ .

We can also estimate qualitatively the phase

volumes corresponding to the "scattering" and decay processes. To this end we neglect the influence of the impurities on the continuous spectrum of the oscillations of the atoms of the host lattice and replace  $\rho_{\alpha\beta}(l\kappa, l'\kappa'; \omega)$  by

$$\rho_{\alpha\beta}^0(l\kappa, l'\kappa'; \omega) = -\text{Im} D_{\alpha\beta}^{(0)R}(l\kappa, l'\kappa', \omega).$$

Then

$$\begin{aligned} \Gamma_1^{(4)} = & \frac{4^2(4-1)!}{a^2} \left( \frac{\hbar}{2M_\kappa a} \right)^3 \left[ \frac{v_0}{(2\pi)^3} \right]^3 \sum_{t, t', t''} \int d^3q d^3q' d^3q'' \\ & \times \frac{|E(0, t\mathbf{q}, t'\mathbf{q}', t''\mathbf{q}'')|^2}{\hbar^2 \omega_{t\mathbf{q}} \omega_{t'\mathbf{q}'} \omega_{t''\mathbf{q}''}} \delta(\omega_{10c} - \omega_{t\mathbf{q}} - \omega_{t'\mathbf{q}'} - \omega_{t''\mathbf{q}''}) \\ & \times [(1 + N_{t\mathbf{q}})(1 + N_{t'\mathbf{q}'})(1 + N_{t''\mathbf{q}''}) - N_{t\mathbf{q}} N_{t'\mathbf{q}'} N_{t''\mathbf{q}''}], \\ \Gamma_0^{(4)} = & \frac{4^2(4-2)! 3}{a^2} \left( \frac{\hbar}{2M_\kappa a} \right)^2 \left( \frac{\hbar}{2M'a} \right) \left[ \frac{v_0}{(2\pi)^3} \right]^2 \\ & \times \sum_{t, t'} \int d^3q d^3q' \frac{|E(0, t\mathbf{q}, t'\mathbf{q}')|^2}{\hbar^2 \omega_{10c} \omega_{t\mathbf{q}} \omega_{t'\mathbf{q}'}} \\ & \times \delta(\omega_{10c} + \omega_{t\mathbf{q}} - \omega_{10c} - \omega_{t'\mathbf{q}'}) [(1 + N_{t'\mathbf{q}'}) N_{t\mathbf{q}}]. \end{aligned} \quad (19)$$

Here  $E(0, t\mathbf{q}, t'\mathbf{q}')/a^4$  and  $E(0, t\mathbf{q}, t'\mathbf{q}', t''\mathbf{q}'')/a^4$  are the Fourier components of the corresponding  $\Phi$  from (16).

It is obvious that for vibrational spectra of typical alkali-halide crystals, the energy conservation law in  $\Gamma_1^{(4)}$  allows only a small region of the phase space near  $\omega_L$ , whereas the contribution to  $\Gamma_0^{(4)}$  is made by the entire region of frequencies. Thus, the ratio of the phase volume can strengthen the inequality (18). Consequently we have

$$\Gamma_0^{(4)} > \Gamma_1^{(4)} \quad (21)$$

up to a temperature  $\Theta_{\text{eff}}$  which is so low, that owing to  $N_{t\mathbf{q}} \ll 1$  the value of  $\Gamma_0^{(4)}$  becomes comparable with  $\Gamma_1^{(4)}$ . We note that  $\Theta_{\text{eff}}$  is lower than the Debye temperature  $\Theta$ . It is obvious that when  $T \ll \Theta$  the value of  $\Gamma_1^{(4)}$  varies with temperature approximately like  $\exp(-\hbar\omega_L/T)$ , whereas  $\Gamma_0^{(4)}$  is proportional to  $(T/\Theta)^7$  at the same temperatures.

Thus, the temperature dependence of the line width of the ion  $H^-$  at temperatures  $T > \Theta_{\text{eff}}$  is determined by the value of  $\Gamma_0^{(4)}$ . In the limit when  $T > \Theta$ , this dependence becomes quadratic. When  $T < \Theta_{\text{eff}}$ , the width is determined by  $\Gamma_1^{(4)}$  and does not depend on the temperature.

For the  $D^-$  ion, the frequency of the local oscillation lies in the range  $\omega_L < \omega_{10c} < 2\omega_L$ . The local peak can in this case become broadened as a result of the third-order anharmonic terms. It

is seen from (14) that the energy conservation law allows in this case only decay into two oscillations of the continuous spectrum. All processes become possible in  $\Gamma_{\alpha\alpha}^{(4)}$  (15). By reasoning similar to that used for the  $H^-$  ion, it can be verified that in this case  $\Gamma_0^{(4)}$  makes the largest contribution to  $\alpha_D(\omega)$ . The experimentally observed temperature dependence of the width of the  $D^-$  peak, intermediate between  $T$  and  $T^2$ , offers evidence that  $\Gamma^{(3)}$  and  $\Gamma^{(4)}$  make a comparable contribution to the line width. As shown in [7], such a situation takes place for the line width of the fundamental absorption by the lattice of pure crystals. However, in order to explain the presence of a temperature dependence of the width of the  $D^-$  peak in the temperature interval  $\Theta_{\text{eff}} < T < \Theta$ , it must be assumed that the quantity  $\Gamma_0^{(4)}$  is not only comparable with but even somewhat larger than  $\Gamma^{(3)}$ .

Let us estimate now the ratio of the line widths for  $H^-$  and  $D^-$  ions. As already noted in Sec. 1, the relative line width is

$$\gamma(\omega_{\text{loc}}) = \frac{1}{2(1-\varepsilon)} D^{(0)R}(0, 0; \omega_{\text{loc}}) \Gamma(0, 0; \omega_{\text{loc}}).$$

We then have from (16) or from (20) and (3)

$$\frac{\gamma_H(\omega_H)}{\gamma_D(\omega_D)} \approx \frac{M_D \omega_D^2 (\Gamma_0^{(4)})_H}{M_H \omega_H^2 (\Gamma_0^{(4)})_D}. \quad (22)$$

Taking into account the approximate relation  $\omega_{\text{loc}} = (\langle \omega^2 \rangle M/M')^{1/2}$ , we obtain finally for the relative widths at temperatures  $T > \Theta_{\text{eff}}$ :

$$\frac{\gamma_H(\omega_H)}{\gamma_D(\omega_D)} \approx \frac{(\Gamma_0^{(4)})_H}{(\Gamma_0^{(4)})_D} \approx \frac{M_D \omega_D}{M_H \omega_H} \approx \left( \frac{M_D}{M_H} \right)^{1/2} > 1. \quad (23)$$

At a temperature  $T < \Theta_{\text{eff}}$ , when  $\Gamma_0^{(4)}$  is negligibly small, the ratio of the widths is determined by the ratio of the quantities  $(\Gamma_1^{(4)})_H$  and  $(\Gamma^{(3)} + \Gamma_1^{(4)})_D$ . In this case the inequality (23) should be reversed.

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**Note added in proof (May 3, 1966).** A recent paper by M. A. Ivanov et al. (FTT, 8, 192, 1966, Soviet Phys. Solid State 8, 150, 1966) is devoted to the investigation of the line width of local oscillations. Our results coincide in part with the results of this paper.

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