

## ON THE CHANGE IN THE FORCE CONSTANTS OF THE IMPURITY ATOMS

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The change in the force constants of the light impurity atoms located at one-dimensional lattice sites is investigated using the pseudoharmonic approximation. A finite change in force constants of impurity atoms and a decrease in frequency of the local modes is obtained for high and low temperature limits. It is assumed that the bare potential of the impurity atoms is the same as that of the host atoms. It is shown that at a sufficiently high temperature the lattice becomes unstable at the impurity lattice site when the impurity mass  $m < 0.28 M$ .

*Introduction*

The introduction of a light impurity atom into a crystal lattice may give rise to additional frequencies of local modes which are greater than the maximum frequency of lattice vibration (cf. *e. g.*, [1]). There are indications from many experimental papers on the spectra of inelastically scattered neutrons [2–5] that the energy of local modes in a crystal differs from the theoretical values calculated for the case of isotopic impurity. This effect is connected with the change in the force constants of the impurity atom.

The present paper discusses the behaviour of a light impurity atom located in a lattice site and the change in its force constants due to strong anharmonicity. It is well known that the amplitude of the impurity atom is greater than the amplitude of host atoms and therefore anharmonic effects appear much earlier in the case of the former.

A method permitting higher-order anharmonic effects to be taken into account, has been developed by Plakida and Siklós [6–8]. The present paper is limited to the case of pseudoharmonic approximation in which the force constants are temperature-dependent and are calculated by means of the self-consistent potential method. The considerations of vibration damping and additional renormalization of phonon energy coming from the decay of phonons would lead to some changes in the values of the parameters obtained here: these factors, will be, however, not considered.

The discussion will be limited to a one-dimensional chain containing a single impurity atom of a mass lower than the host atoms and having the same bare potential as the latter.

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Interactions of nearest neighbours only are taken into account. The consideration of anharmonicity leads to renormalization of energy of local modes, to a change in interatomic distances in the neighbourhood of the impurity atom and to the instability of the impurity atom in a certain temperature range.

### 1. Pseudoharmonic approximation

The pseudoharmonic approximation takes into account the renormalization of phonon energy in the self-consistent field of all other phonons. This approximation corresponds to the first-order perturbation theory for the mass operator in which, however, the contribution of anaharmonic terms of all orders are taken into account. In contrast to the harmonic approximation, the force constants in this approximation are second-order derivatives of self-consistent potential which can be obtained by averaging the bare potential with the function  $e^{-x^2/2}$  over the region of mean displacements of atoms from their equilibrium positions.

The frequency spectrum of atomic vibrations in a one-dimensional lattice containing impurity atoms is given by poles of the Green's function  $G_{mn}(\omega)$  which in the pseudoharmonic approximation can be found by solving the following system of equations [6-8].

$$\sum_m \{M_m \omega^2 \delta_{nm} - F_{nm}\} G_{mn'}(\omega) = \delta_{nn'} \quad (1.1)$$

$$F_{nm} = \begin{cases} \left. \frac{\partial \tilde{\Phi}(R_n - R_m)}{\partial R_n \partial R_m} \right|_{\substack{R_n = n \\ R_m = m}} & \text{where } n \neq m \\ - \sum_{m' \neq n} F_{nm'} & \text{where } n = m \end{cases} \quad (1.2)$$

$$\tilde{\Phi}(R_n - R_m) = \langle \Phi(R_n - R_m) \rangle = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} dx e^{-\frac{x^2}{2}} \Phi(R_n - R_m + x \sqrt{\langle (u_n - u_m)^2 \rangle}) \quad (1.3)$$

$$\langle (u_n - u_m)^2 \rangle = \langle u_n^2 \rangle + \langle u_m^2 \rangle - 2\langle u_n u_m \rangle \quad (1.4)$$

$$\langle u_n u_m \rangle = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega}{e^{\frac{\omega}{\theta}} - 1} \{-2 \operatorname{Im} G_{mn}(\omega + i\eta)\} \quad (1.5)$$

where  $M_m$  is the mass of the  $m$ -th atom,  $F_{nm}$  is the force constant, and  $\langle (u_n - u_m)^2 \rangle$  is the mean square relative displacement.

The system of equations (1.1) through (1.5) is self-consistent, the calculation of the potential (1.3) requires the knowledge of the mean square displacement (1.4) and (1.5) which is dependent on the lattice spectrum (1.1) which in turn depends on the force constants (1.2).

To facilitate the calculations the author has accepted the Morse potential [9] as bare interaction potential.

$$\Phi(R) = D_0 \{(e^{-2a_0(R-r_0)} - 1)^2 - 1\} \quad (1.6)$$

where  $D_0$  is the depth of the potential well,  $r_0$  the mean distance between atoms at  $0^\circ\text{K}$ , and  $\frac{1}{a_0}$  the width of the well.

The self-consistent Morse potential calculated by means of Eq. (1.3) has the following form

$$\tilde{\Phi}_{nm}(R) = D_0 \left\{ e^{-2a_0(R-r_0)} e^{2a_0\langle u_n - u_m \rangle} - 2e^{-a_0(R-r_0)} e^{\frac{a_0^2}{2}\langle (u_n - u_m)^2 \rangle} \right\} \quad (1.7)$$

### 2. One-dimensional lattice containing no impurity atoms

We shall consider a linear chain with interaction of nearest neighbours. By making use of Eqs. (1.1) through (1.7) and the fact that the Green's function depends only on the difference of the indices ( $m-n$ ), we obtain the following system of equations (cf. [7, 8]):

$$\omega_k^2(\Theta) = \omega_L^2(\Theta) \sin^2 \frac{kl_0}{2} \quad (2.1)$$

$$\omega_L^2(\Theta) = \frac{4F(\Theta)}{M} \quad (2.2)$$

$$F(\Theta) = D_0 a_0^2 e^{-a_0^2 \bar{u}^2} \quad (2.3)$$

$$\bar{u}_0^2 = \langle (u_n - u_{n-1})^2 \rangle = \frac{1}{2NF(\Theta)} \sum_k \omega_k(\Theta) \operatorname{cth} \frac{\omega_k(\Theta)}{2\Theta} \quad (2.4)$$

where  $l_0$  is the mean distance between nearest neighbours which is determined from the condition that the first derivative of the self-consistent potential (1.3) should vanish. We then obtain for zero external pressure

$$l_0 = r_0 + \frac{3}{2} a_0 \bar{u}_0^2 \quad (2.5)$$

The system of equations (2.1) through (2.4) permits the mean square displacement  $\bar{u}_0^2$  to be found as a function of temperature and thus enables us to calculate the temperature dependence of the force constant (2.3), the spectrum (2.1) and the interatomic distance  $l_0$ .

The Fourier transform of the Green's function of this system has a simple form (cf. [7, 8])

$$g_{mn}(\omega) = \frac{1}{NM} \sum_k e^{ikl_0(m-n)} \frac{1}{\omega^2 - \omega_k^2(\Theta)} = \frac{1}{NM} \sum_k \cos kl_0(m-n) \frac{1}{\omega^2 - \omega_k(\Theta)}. \quad (2.6)$$

### 3. One-dimensional lattice containing an impurity atom

Suppose that an atom located at a site  $n = 0$  is replaced by another atom of a smaller mass. We shall assume that the interaction of the impurity atom with other atoms has the form of Morse potential (1.7). The mean distance between the impurity and the nearest neighbour is (like in the case of an ideal lattice)

$$l = r_0 + \frac{3}{2} a_0 \bar{u}^2 \quad (3.1)$$

where  $\bar{u}^2 = \langle (u_0 - u_1)^2 \rangle$  is the mean square displacement of the impurity atom with respect to its neighbour.

The force constant between the impurity atom and its neighbour is the second derivative of the self-consistent potential (1.7) calculated at the position  $l$  Eq. (3.1)

$$f(\Theta) = D_0 a_0^2 e^{-a_0^2 \bar{u}^2} \quad (3.2)$$

The difference in the values of the mean vibration amplitude of the impurity atom and the mean vibration amplitude of the host atoms results from the mass difference and different distances between the atoms ( $l \neq l_0$ ). This leads to another value of the force constant between the impurity atom and its nearest neighbour (see Eq. 3.2).

In principle it is possible to find  $f(\Theta)$  by solving the system of equations (1.1) through (1.4), it is, however, much simpler to find its relative change

$$\gamma(\Theta) = \frac{f(\Theta) - F(\Theta)}{F(\Theta)} \quad (3.3)$$

We therefore divide Eq. (3.2) by Eq. (2.3) and obtain

$$1 + \gamma(\Theta) = e^{-a_0^2 \{\bar{u}^2(\epsilon, \gamma) - \bar{u}_0^2\}} \quad (3.4)$$

Eq. (3.4) is a transcendental equation with respect to  $\gamma(\Theta)$ . To solve this equation it is necessary to find the explicit form of the dependence of the mean square relative displacement of the impurity atom  $\bar{u}^2$  on  $\gamma(\Theta)$ .

The temperature-dependent deformation of the lattice is given by

$$l - l_0 = -\frac{3}{2a_0} \ln(1 + \gamma(\Theta)) \quad (3.5)$$

where  $\gamma(\Theta)$  is the solution of Eq. (3.4).

#### 4. Mean square displacement of impurity atom vibration

Our problem is to find the Green's function for a lattice containing an impurity atom and to express it in terms of the Green's functions of the host crystal (2.6).

The equation for the Green's function in the case of a lattice containing impurity atoms in pseudoharmonic approximation can be written according to Eq. (1.1) in matrix form

$$\bar{L} \cdot \bar{G} = \bar{I} \quad (4.1)$$

For crystals with no impurities we have

$$\bar{L}^0 \cdot \bar{g} = \bar{I} \quad (4.2)$$

where

$$L_{nm}^0 = M\omega^2 \delta_{nm} - F_{nm} \quad (4.3)$$

and  $g_{mn}$  stands for the Green's function (2.6).

The elements of the  $L$  matrix can be written in the form

$$L_{nm} = L_{nm}^0 - \lambda L_{nm} \quad (4.4)$$

where

$$\lambda L_{nm} = \varepsilon M \omega^2 \delta_{nm} - (F_{nm} - f_{nm})$$

and  $\varepsilon = 1 - \frac{m}{M}$  is the mass defect.

Assuming that the introduced impurity changes the force constants of the nearest neighbours only, we can write the perturbation matrix in the form

$$\bar{\delta L} = \begin{vmatrix} \lambda_0 + 2\gamma F, & -\gamma F, & -\gamma F, & 0, & \dots \\ -\gamma F, & \gamma F, & 0, & 0, & \dots \\ -\gamma F, & 0, & \gamma F, & 0, & \dots \\ 0, & 0, & 0, & 0, & \dots \\ \dots & \dots & \dots & \dots & \dots \end{vmatrix} \quad (4.5)$$

where  $\lambda_0 = M \omega^2 \varepsilon$ . Taking into account Eqs. (4.1), (4.2) and (4.4) we find the Green's function for the crystal containing the impurity:

$$\bar{G} = (\bar{1} - \bar{g} \cdot \bar{\delta L})^{-1} \cdot \bar{g} \quad (4.6)$$

In the following we shall be interested only in the Green's functions corresponding to the sites [00], [01] and [11]. It is therefore possible to discuss the solution of Eq. (4.6) only for a 3-rd order matrix which describes the impurity atom and its neighbours. We introduce a unitary matrix.

$$T = \begin{vmatrix} 1 & 0 & 0 \\ 0 & \frac{1}{\sqrt{2}} & -\frac{1}{\sqrt{2}} \\ 0 & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} \end{vmatrix} \quad (4.7)$$

and make the following transformation

$$T^T \cdot G \cdot T = [T^T \cdot (\bar{1} - \bar{g} \cdot \bar{\delta L})^{-1} \cdot T \cdot T^T \cdot \bar{g} \cdot T] \quad (4.8)$$

The matrix  $M(\omega^2) = T^T \cdot (\bar{1} - \bar{g} \cdot \bar{\delta L}) \cdot T$  will have a quasidiagonal form. The condition of vanishing determinant of this matrix defines the poles of the Green function (4.6), *i.e.* the characteristic frequencies of the vibration spectrum of our lattice. Taking into account the properties of the corresponding determinant we obtain:

$$\det M(\omega^2) = \det \begin{vmatrix} M_s(\omega^2) & 0 \\ 0 & M_p(\omega^2) \end{vmatrix} = \det M_s(\omega^2) \cdot \det M_p(\omega^2) = 0 \quad (4.9)$$

where

$$\begin{aligned}\det M_s(\omega^2) &= 1 + \gamma - M\omega^2[(\varepsilon + \gamma)g_{00} + \gamma(\varepsilon - 1)g_{01}] \\ \det M_p(\omega^2) &= 1 - \gamma F [g_{11} - g_{1\bar{1}}]\end{aligned}\quad (4.10)$$

In the derivation of (4.10) we have made use of the properties of the Green's function for a one-dimensional lattice (see Ref. [11]):

$$\begin{aligned}g_{00} - g_{10} &= \frac{1}{2F} (M\omega^2 g_{00} - 1) \\ 2g_{10} - g_{11} - g_{1\bar{1}} &= \frac{M}{F} \omega^2 g_{01} \\ 2g_{10}g_{01} - g_{00}(g_{11} + g_{1\bar{1}}) &= \frac{1}{F} g_{01}.\end{aligned}\quad (4.11)$$

Owing to the transformation (4.8) we succeeded in separating two types of vibrations: vibrations of the  $s$  type for which the essential point is the change in the mass; this is a localized mode in which the main role is played by the impurity atom. The other type,  $p$  is due to the change in the force constants between the impurity and the neighbouring atom. The impurity does not participate in vibrations of the  $p$ -type.

We can readily find the inverse matrix  $M^{-1}(\omega^2)$  and then the Green's function matrix for the lattice containing the impurity  $T^T \cdot G \cdot T$ . In order to calculate the mean square relative displacement of the impurity atom it is necessary to express  $\text{Im}(G_{00} + G_{11} - 2G_{01})$  in terms of the  $T^T \cdot G \cdot T$  matrix elements with the help of the identity

$$\text{Im}(G_{00} + G_{11} - 2G_{01}) = \text{Im} \left[ G_{00} + \frac{1}{2} (G_{11} + G_{1\bar{1}}) + \frac{1}{2} (G_{11} - G_{1\bar{1}}) - 2G_{01} \right] \quad (4.12)$$

The imaginary parts of the matrix elements of  $T^T \cdot G \cdot T$  are found by decomposing both the numerator and the denominator of each matrix element, (*i.e.*, the Green functions (2.6)) into the real and imaginary part. The expression of the type (4.1) obtained in this way is substituted into the spectral theorem (1.4), (1.5).

Hence we obtain for the mean square relative displacement

$$\begin{aligned}\bar{u}^2 &= \frac{1}{2NF} \sum_k \omega_k \text{cth} \left\{ \frac{\omega_k}{2\Theta} \left[ \frac{S(\omega_k^2)}{V_1^2(\omega_k^2) + V_2^2(\omega_k^2)} + \right. \right. \\ &\left. \left. + \frac{P_k}{W_1^2(\omega_k^2) + W_2^2(\omega_k^2)} \right] \right\} + \frac{1}{2\omega_{\text{loc}}} \text{cth} \left\{ \frac{\omega_{\text{loc}}}{2\Theta} \left[ \frac{R(\omega_{\text{loc}}^2)}{\frac{dV_1}{d\omega^2}(\omega^2 = \omega_{\text{loc}}^2)} \right] \right\}\end{aligned}\quad (4.13)$$

where

$$\begin{aligned}S(\omega_k^2) &= \frac{1}{2} [1 - \varepsilon - \cos kl_0(1 + 2\gamma - \varepsilon)] \\ V_1(\omega^2) &= \begin{cases} 1 + \gamma & \text{for } 0 \leq \omega^2 \leq \omega_L \\ (1 + \gamma) - (\varepsilon + \gamma) \sqrt{\frac{\omega^2}{\omega^2 - \omega_L^2}} & \text{for } \omega^2 > \omega_L \end{cases}\end{aligned}$$

$$V_2(\omega^2) = (\text{sign } \omega) (\varepsilon + \gamma) \sqrt{\frac{\omega^2}{\omega_L - \omega^2}} \quad \text{for } 0 \leq \omega^2 \leq \omega_L \quad (4.14)$$

$$R(\omega_{\text{loc}}) = \frac{1}{F\omega_L} \left( \omega_{\text{loc}} + \frac{\omega_L}{2} \right) \left( \frac{\omega_{\text{loc}}}{\sqrt{\omega_{\text{loc}}(\omega_{\text{loc}} - \omega_L)}} - 1 \right) + \\ + \frac{(\varepsilon - 1)\omega_{\text{loc}}}{2F\sqrt{\omega_{\text{loc}}(\omega_{\text{loc}} - \omega_L)}} \quad \text{for } \omega_{\text{loc}} > \omega_L \quad (4.15)$$

$$P_k = \frac{1}{2} (1 + \cos kl_0)$$

$$W_1(\omega^2) = 1 + \gamma \left( 1 - \frac{2\omega_k}{\omega_L} \right) \quad \text{for } 0 \leq \omega^2 \leq \omega_L$$

$$W_2(\omega^2) = \frac{2(\text{sign } \omega)}{\omega_L} \gamma \omega \sqrt{\omega_L - \omega^2} \quad \text{for } 0 \leq \omega^2 \leq \omega_L \quad (4.16)$$

It was assumed in the derivation of Eqs (4.14) through (4.16) that the mass of the impurity atom is smaller than that of the host atoms ( $m < M$ ) and that the terms of the order of  $\gamma \frac{m}{M}$  are negligible. This is a good approximation for small masses of the impurity atoms and gives also a proper result for  $m \rightarrow M$  since in this case  $\gamma \rightarrow 0$ .

The first term of Eq. (4.13) describes a system of vibrations of the  $s$ -type, the second the  $p$ -type, while the last one takes into account local  $s$ -type modes which are induced at frequencies greater than the maximum frequency  $\omega_L$ . The frequency of local modes is defined by the equation  $V_1(\omega^2) = 0$  and in our approximation is equal

$$\omega_{\text{loc}} = \frac{\omega_L}{\sqrt{1 - \left( \frac{\varepsilon + \gamma}{1 + \gamma} \right)^2}} \quad (4.17)$$

for  $|\gamma| < \varepsilon$ .

If the relative change in the force constants exceeds the mass defect then local mode frequency goes over into continuous spectrum.

Substituting Eqs (4.14) through (4.16) as well as the value of the local frequency (4.17) into the formula for  $\bar{u}^2$  (4.13) we can calculate the mean square displacement for two limiting cases  $\Theta \gg \omega_L$  or  $\Theta \rightarrow 0$ .

At high temperatures at which  $\omega_L \gg \Theta$  and  $\text{cth } x \approx 1/x$ , we can write

$$\bar{u}^2 - \bar{u}_0^2 = \frac{\Theta}{F} [L_1(\varepsilon, \gamma) + s_1(\varepsilon, \gamma) + p_1(\gamma) - 1] \quad (4.18)$$

while at low temperatures ( $\omega_L \gg \Theta$  and  $\text{cth } x \approx 1$ ).

$$\bar{u}^2 - \bar{u}_0^2 = \frac{\omega_L(\Theta)}{\pi F(\Theta)} [L_0(\varepsilon, \gamma) + s_0(\varepsilon, \gamma) + p_0(\gamma) - 1] \quad (4.19)$$

The functions  $s_1, s_0, p_1, p_0, L_1, L_0$  describe the contribution of the  $s$ -type,  $p$ -type, and local modes, respectively, and are given in Ref. [12].

They can be easily obtained by replacing the sum over  $k$  in Eq. (4.13) by an integral and taking into account the approximation for the function  $\text{cth } x$ . In the derivation of (4.18) and (4.19) we have made use of the approximation  $\bar{u}_0^2 = \frac{\Theta}{f}$  for  $\omega_L \ll \Theta$  and  $\bar{u}_0^2 = \frac{\omega}{\pi F(\Theta)}$  for  $\Theta \ll \omega_L$ .

The ratio of the vibration amplitudes can be obtained from Eqs (4.18) and (4.19) in the form

$$\sqrt{\frac{\bar{u}^2}{\bar{u}_0^2}} = \sqrt{L(\varepsilon, \gamma) + s(\varepsilon, \gamma) + p(\gamma)} \quad (4.20)$$

The relative distance between the impurity atom and its nearest neighbour is according to Eqs (2.5) and (3.6)

$$\frac{l}{l_0} = 1 - \frac{\frac{3}{2} \ln(1+\gamma)}{r_0 a_0 + \frac{3}{2} \bar{u}_0^2} \quad (4.21)$$

### 5. Solution of the self-consistent equation

#### A. High temperatures ( $\omega_L \ll \Theta$ )

By taking into account the first term of the expansion of  $\text{cth } \frac{\omega_L}{2\Theta} \approx \frac{2\Theta}{\omega_k}$  in Eq. (2.4) and making use of Eq. (2.3) we obtain self-consistent equations for a pure linear lattice (*cf.* Refs [7] and [8])

$$\frac{D_0}{\Theta} y_\Theta = e^{y_\Theta} \quad (5.1)$$

where

$$y_\Theta = a_0^2 \bar{u}_0^2 = - \frac{a_0^2 \Theta}{F(\Theta)} \quad (5.2)$$

The preceding equation has a solution at  $\Theta < \Theta_c = \frac{D_0}{e}$ . For temperatures higher than the critical  $\Theta_c$ ,  $\bar{u}^2$  is complex and the lattice becomes unstable.

The parameter  $\gamma$ , which describes the change in the force constants of the impurity atom, is the solution of the equation

$$- \frac{F(\Theta)}{\Theta a_0^2} \ln(1+\gamma) = L_1(\varepsilon, \gamma) + s_1(\varepsilon, \gamma) + p_1(\gamma) - 1 \quad (5.3)$$

which results from the expressions (3.4) and (4.8).



Fig. 1 shows the solution of Eq. (5.3) as a function of the temperature for a few values of the mass defect. The temperature change in the force constant  $F(\Theta)$  of the host lattice has been taken into account in this calculation. It is seen that the relative change in the

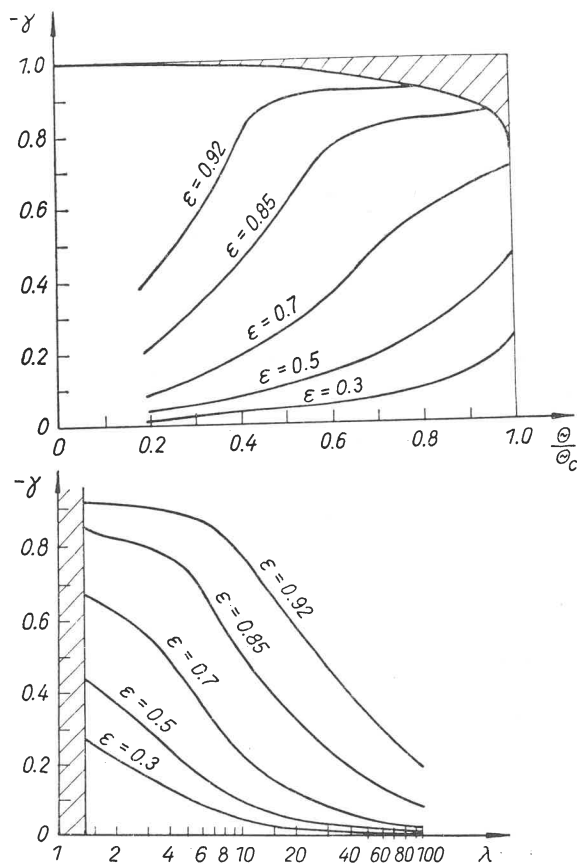


Fig. 1. Solution of self-consistent equations (5.3) and (5.8): The dependence of the change  $\gamma$  in the force constants of the impurity atom on temperature  $\Theta$  (for  $\omega_L \ll \Theta$ ), and on  $\lambda = \frac{\pi \sqrt{D_0 M}}{2a_0}$  (for  $\omega_L \gg \Theta$ ).  $\Theta_c$  is the critical temperature of instability of host lattice. Hatched area corresponds to instability region. For  $\omega_L \ll \Theta$  (upper part of the figure) the instability occurs at the site of the impurity; for  $\omega_L \gg \Theta$  (bottom part) the hatched area corresponds to instability of lattice as a whole

force constants of the impurity atom increases with increasing temperature. For very light impurities ( $\epsilon > \epsilon_{cr}$ ) the lattice becomes unstable at the position of the impurity atom for a smaller temperature than the critical temperature of the host lattice. At the critical temperature  $\Theta_{cr}$  the change in the force constants is equal to the mass defect  $\gamma = -\epsilon$ . The value  $\Theta_{cr}$  can be found from the following equation

$$\frac{a_0^3 \Theta_{cr}}{F(\Theta_{cr})} = -\frac{2}{3} (1-\epsilon) \ln (1-\epsilon) \quad (5.4)$$

For critical values of the mass defect the critical temperatures of the impurity and the host lattice are the same  $\Theta_{cr} = \Theta_c$ . By making use of this condition we can find the numerical value of  $\varepsilon_{cr} = 0.72$ . For  $\varepsilon < \varepsilon_{cr}$  the crystal containing the impurity is stable up to the critical temperature of the host lattice  $\Theta_c$ .

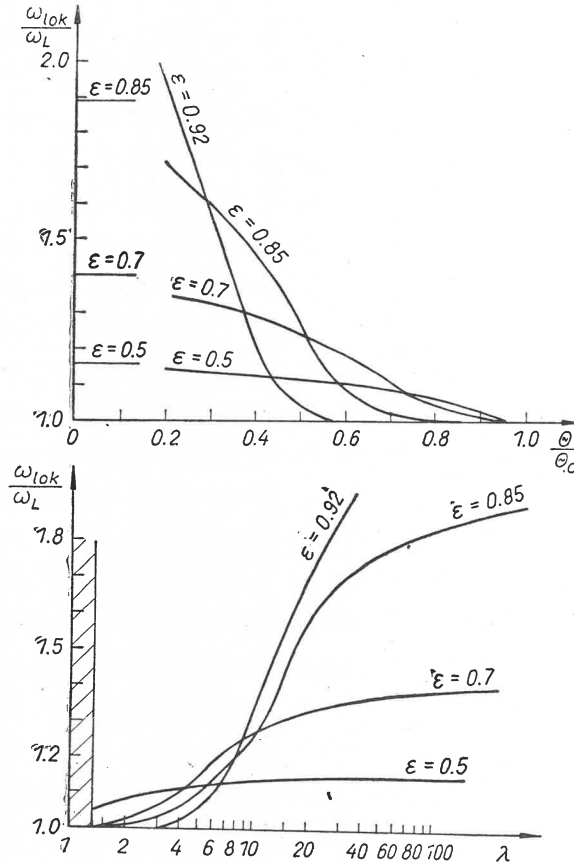


Fig. 2. Dependence of local mode frequency on temperature (for  $\omega_L \ll \Theta$ ), and on  $\lambda = \frac{\pi \sqrt{D_0 M}}{2a_0}$  (for  $\omega_L \gg \Theta$ ).  $\Theta_c$  is the critical temperature of instability of host lattice. The local mode frequencies of isotopic impurity are marked on vertical axis

The frequency of the local mode (Fig. 2) decreases with increasing temperature. The distance between the impurity and its nearest neighbours Eq. (4.2) is shown in Fig. 3. It should be pointed out that the ends of the lattice in consideration are free and thus there are no additional forces counteracting its dilatation.

The amplitude of the impurity atom  $\sqrt{u^2}$  is about 1.5–3 times as large as the amplitude of host atoms and amounts to about  $\frac{1}{10} \div \frac{1}{3}$  of the interatomic distance  $l_0$ .

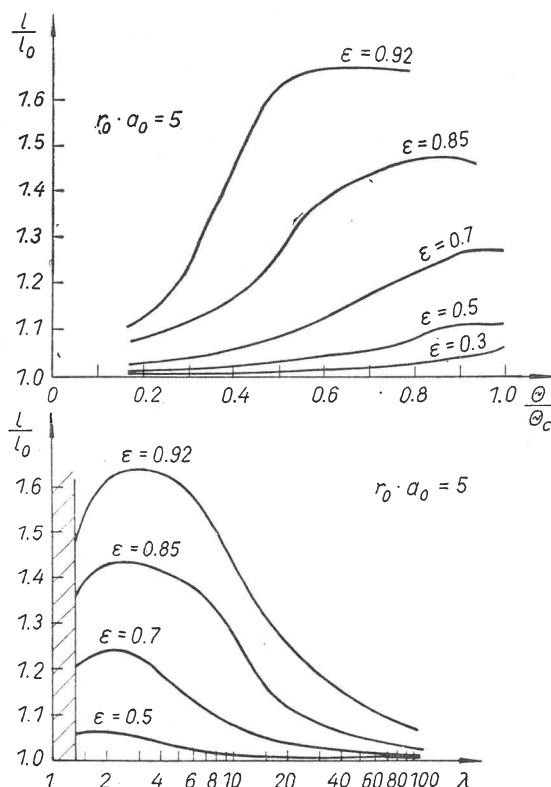


Fig. 3. Dependence of the distance between the impurity and its nearest neighbour on temperature  $\Theta$  (for  $\omega_L \ll \Theta$ ), and on  $\lambda = \frac{\pi \sqrt{D_0 M}}{2a_0}$  (for  $\omega_L \gg \Theta$ ).  $\Theta_c$  is the critical temperature of instability of the host lattice

### B. Low temperatures ( $\omega_L \gg \Theta$ )

The system of equations (2.1) through (2.4) has in this case the form (cf. Refs [7, 8])

$$\lambda^2 y_\lambda^2 = e^{y_\lambda} \quad (5.5)$$

where

$$\lambda = \frac{\pi \sqrt{M D_0}}{2a_0} \quad (5.6)$$

and

$$y_\lambda = a_0^2 u_0^2 = \frac{a_0^2 \omega_L(\Theta)}{\pi F(\Theta)} \quad (5.7)$$

If the parameters of the lattice are such that  $\lambda < \lambda_c = e/2$  then Eq. (5.5) will have no real solutions and the lattice will be unstable even for  $\Theta = 0^\circ\text{K}$ .

In accordance with Eqs. (3.5) and (4.21) the low-temperature behaviour of the impurity is given by the equation

$$-\frac{1}{y_\lambda} \ln(1 + \gamma) = L_0(\epsilon, \gamma) + s_0(\epsilon, \gamma) + p_0(\gamma) - 1. \quad (5.8)$$

where  $\gamma_\lambda$  is the solution of Eq. (5.5) for a given value of  $\lambda$ . The solution  $\gamma$  of this equation (5.8) is shown in Fig. 1. The impurity remains in this case stable.

When increasing the parameter  $\lambda$  the frequency of the local mode (Fig. 2) approaches its limiting value *i.e.* the frequency of the local mode of the isotopic impurity. The distance between the impurity and its nearest neighbour (Fig. 3) decreases with increasing  $\lambda$ . At the temperature of 0°K the amplitude of the impurity atom vibration  $\sqrt{u^2}$  is about  $2 \div 4$  times as large as that of the host atoms  $\sqrt{u_0^2}$  and amounts to about  $\frac{1}{10} \div \frac{1}{3}$  of the interatomic distance  $l_0$ .

## 6. Conclusions

By making use of the pseudoharmonic approximation we have calculated the decrease in the force constants of the impurity atom for zero external pressure. This change results mainly from the change in the mean internatomic distance (3.5). The change in the distance between the impurity and its nearest neighbour is due to the larger amplitude of the impurity. The change in the force constants gives rise to a change in the frequencies of local modes.

It is shown that a very light impurity ( $\varepsilon > \varepsilon_{cr}$ ) is unstable in some temperature range. The amplitude of the impurity atom is always smaller than the lattice constant.

The consideration of the decay of phonons would lead to additional changes in the force constants of the impurity, renormalization of the local mode frequency, vibration damping and would also affect the stability conditions of the impurity.

In the case of a three-dimensional crystal the change in interatomic distances is much more difficult and, therefore, we cannot expect that the changes in the force constants and local mode frequency in real crystals will be as large as in the case of the linear crystal under consideration. The stability conditions of the impurity will be, however, more severe and thus more difficult to be satisfied. We should also bear in mind that the bare potential of the impurity atoms may be different from the bare potential of host atoms.

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