THE MÖSSBAUER·EFFECT FOR AN IMPURITY NUCLEUS IN A CRYSTAL. II

Yu. KAGAN and Ya. IOSILEVSKIĬ

Submitted to JETP editor July 31, 1962

J. Exptl. Theoret. Phys. (U.S.S.R.) **44**, 284—302 (January, 1963)

Using the method developed earlier, [1] we treat the resonance absorption of γ quanta by an arbitrary impurity nucleus, accompanied by a change in the state of the macroscopic system. The relation of the probability for one-quantum excitation to the vibration spectrum of the crystal is analyzed. The intensity corresponding to the excitation of localized levels is determined. A detailed analysis is given of the temperature dependence of the Mössbauer effect and of one-quantum excitations, taking into account, in particular, the effect of degeneracy. Computational results are presented for the case of a host lattice with a simple unit cell, including nearest neighbor interactions. A comparison of the theoretical and experimental results is given.

1. INTRODUCTION

IN an earlier paper [1](to be cited as I), the authors developed a theory describing the Mössbauer effect on a nucleus which is different from the nuclei of the host lattice. For monatomic lattices with cubic symmetry, explicit expressions were given for the probability of the effect for any ratio of masses of the impurity nucleus and the nuclei of the host lattice.

The present paper is a direct continuation of I. It presents, in addition to a detailed analysis of resonance absorption (emission) of γ quanta with no change in the energy of the macroscopic system, a treatment of transitions accompanied by a change in the state of the system. The main interest is centered on one-quantum transitions, since the intensity of the corresponding γ radiation gives information about the nature of the vibrations of the impurity atom and the host lattice.

There are two problems of primary interest: the study of the localized vibrations, caused by the radiating impurity itself (qualitative arguments about the possibility of obtaining such information from the Mössbauer effect were first given by Maleev [2]), and the possibility of determining the distribution function for the frequencies of an arbitrary regular lattice.

Section 2 gives the general formulas describing elastic (Mössbauer effect) and inelastic processes in an arbitrary, harmonically interacting system of particles when γ emission occurs. Both the quasicontinuous and discrete spectra are treated, over the whole range of temperatures. Section 3 gives a detailed treatment of the case of an isolated impurity nucleus. Section 4 contains a discussion of the results for the Mössbauer effect and for one-quantum excitations in a crystal when there is resonance absorption (emission) by an impurity nucleus. Finally, Sec. 5 gives the results of direct computations based on a model of the crystal lattice which includes nearest neighbor interactions of both central and noncentral type. In this section, a comparison of the theoretical and experimental results is given.

2. GENERAL FORMULAS

Let us consider an arbitrary system of harmonically interacting particles, and let one of the particles undergo resonance absorption (emission) of a γ quantum. We shall assume that the vibration spectrum of the system consists of quasicontinuous bands, whose width Δ is much greater than the width Γ of the nuclear resonance level, and of discrete frequencies which correspond to localized vibrations.

We introduce the reduced displacements l labels the particles) [1]

$$v^i(l) = m_l^{1/2} u^i(l) \tag{2.1}$$

and expand $v^i(l)$ in the dimensionless normal coordinates q_β:

$$v^i(l) = \sum_\beta \sqrt{\frac{\hbar}{\omega_\beta}} S^i(l; \beta) q_\beta. \tag{2.2}$$

The quantities $S^i(l; \beta)$, which are the real and normalized solutions of the equation

$$\omega_\beta^2 S^i(l; \beta) = \sum_{l'} \overline{A}^{ik}(l, l') S^k(l'; \beta), \tag{2.3}$$

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1) Unless otherwise stated, the notation is the same as in I.

form a unitary (orthogonal) matrix. Here

$$A^{ik}(l, l') = (m_l m_{l'})^{-1/2} A^{ik}(l, l'), \qquad (2.4)$$

while $A^{ik}(l, l')$ is the usual coefficient in the quadratic term of the expansion of the potential energy of the system in the displacements.

If, on emission of a γ quantum, the nucleus $l = 0$ receives a momentum $\hbar k$, the probability of a transition of the system with a change $m_\beta = n'_\beta - n_\beta$ in the occupation numbers can be represented as

$$W(\{m_\beta\})$$
$$= \prod_\beta \overline{|\langle n_\beta + m_\beta | \exp [i\sqrt{\hbar/m_0\omega_\beta}\, kS(0; \beta)\, q_\beta] | n_\beta\rangle|^2}, \qquad (2.5)$$

where the dash denotes an average over the initial equilibrium state.

Let us write (2.5) as a product

$$W(\{m_\beta\}) = W_1(\{m_p\})\, W_2(\{m_d\}),$$

where W_1 and W_2 are the respective transition probabilities for the quasicontinuous and the discrete spectrum.

Remembering that the displacement amplitudes in the quasicontinuous spectrum are small, we find for W_1

$$W_1(\{m_p\}) = e^{-Z'} \prod_p \Big\{ \delta_{m_p,0} + \frac{\hbar}{2m_0\omega_p}(kS(0; p))^2 [(\bar n_p + 1)\delta_{m_p, 1}$$
$$+ \bar n_p \delta_{m_p, -1}]\Big\},$$

$$Z' = \frac{\hbar}{2m_0} \sum_p \frac{(kS(0; p))^2}{\omega_p}(2\bar n_p + 1). \qquad (2.6)$$

As was pointed out in I, in computing the matrix elements in (2.5) for the discrete frequencies one must use the exact expression, since the displacement amplitude no longer has the factor $1/N^{1/2}$. We shall use the relation obtained by Bloch and Nordsieck.[3] One can show that the series appearing in [3] reduce to associated Laguerre polynomials $L_n^m(x)$. The general expression for the matrix element can then be written as follows:

$$\langle n + m | \exp [i\sqrt{2x}q] | n\rangle$$
$$= \begin{cases} e^{-x/2}(i\sqrt{x})^m \sqrt{n!/(n+m)!}\, L_n^m(x), & m > 0 \\ e^{-x/2}(i\sqrt{x})^{|m|} \sqrt{(n+m)!/n!}\, L_{n+m}^{|m|}(x), & m \leqslant 0. \end{cases} \qquad (2.7)$$

We note that the series appearing in formula (3.24) of I and corresponding to $m_d = 0$, is precisely the same as L_{n_d}.

Using the well-known relation from the theory of the Laguerre polynomials (cf., for example, [4])

$$(1 - z) \sum_{n=0}^\infty \frac{n!}{\Gamma(n+m+1)}[L_n^m(x)]^2 z^n$$
$$= \exp\Big(-2x\frac{z}{1-z}\Big)x^{-m}z^{-m/2}I_m\Big(2x\frac{z^{1/2}}{1-z}\Big), \quad |z| < 1,$$

where I_m is a Bessel function of pure imaginary argument. Then, averaging the square of (2.7) over the initial state of thermal equilibrium, we find

$$W_2(\{m_d\}) = \exp\Big\{-Z'' + \frac{\hbar}{2kT}\sum_d m_d\omega_d\Big\}$$
$$\times \prod_d I_{m_d}\Big[\frac{\hbar(kS(0; d))^2}{2m_0\omega_d} \text{sh}^{-1}\frac{\hbar\omega_d}{2kT}\Big];$$
$$Z'' = \frac{\hbar}{2m_0}\sum_d \frac{(kS(0; d))^2}{\omega_d}(2\bar n_d + 1). \qquad (2.8)^*$$

In particular, for the probability of a transition which is not accompanied by any change in the system, we have from (2.6) and (2.8)

$$W^0 = W_1^0 W_2^0 = e^{-Z}\prod_d I_0\Big[\frac{\hbar(kS(0; d))^2}{2m_0\omega_d}\text{sh}^{-1}\frac{\hbar\omega_d}{2kT}\Big],$$
$$Z = Z' + Z''. \qquad (2.9)$$

Expression (2.9) coincides with the one found in I, if we use for $S(0, d)$ the appropriate solution of (2.3).[2]

In studying the inelastic processes which accompany resonance absorption or emission of γ quanta, we shall restrict our treatment to cases where $R_0/\hbar\omega_0 < 1$ ($R_0 = \hbar^2 k^2/2m_0$ is the recoil energy for the free nucleus, and ω_0 is a characteristic frequency for the spectrum of the macroscopic system). For a range ΔE of variation of the γ ray energy which is restricted to be of order $\hbar\omega_0$, one-phonon processes play the dominant role. But the relative probability of such processes as a function of ΔE actually gives the maximum of information about the vibration spectrum of the macroscopic system.

First consider one-phonon processes in the quasicontinuous spectrum. For the probability density for emission of a γ quantum with energy E in the direction $\kappa = k/k$, we find, using (2.6) (cf. [5]):

$$\rho^1(\Delta E, \kappa) = WR_0\sum_p \frac{(\kappa S(0; p))^2}{\hbar\omega_p}\Big(\bar n_p + \frac{1}{2} \pm \frac{1}{2}\Big)\delta(\Delta E \mp \hbar\omega_p), \qquad (2.10)$$

$$W = W_1^0 W_2 = e^{-Z}\prod_d' I_0\Big[\sum_{(d)}\frac{\hbar(kS(0;d))^2}{2m_0\omega_d}\text{sh}^{-1}\frac{\hbar\omega_d}{2kT}\Big], (2.10')$$

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*sh = sinh.

[2] The fact that the series in formula (3.25) of I should transform into I_0, to within an exponential factor, was first pointed out to us by V. I. Peresada.

where Π'_d denotes a product for the various discrete frequencies, while $\sum_{(d)}$ is a summation over all the normal modes corresponding to the degenerate discrete frequency ω_d; E_0 is the energy of the excited state of the nucleus ($|\Delta E| \gg \Gamma$), $\Delta E = E_0 - E$.

In all those cases where the surroundings of the radiating nucleus have cubic symmetry, and also when measurements are made for three mutually perpendicular directions κ (for three orientations of the sample corresponding to cyclic permutation of the coordinates), or if one uses a polycrystalline sample and can neglect the anisotropy of W (cf.[5]), one can eliminate the dependence on the direction of the polarization vectors in (2.10). Changing from summation to integration, we get

$$\overline{\left(\frac{\rho^1(\Delta E, \varkappa)}{W}\right)}^{\varkappa} = \frac{NR_0}{\hbar|\Delta E|}\overline{|S(0; p)|^2}_{\hbar\omega_p=|\Delta E|}\,\psi\left(\frac{|\Delta E|}{\hbar}\right)$$
$$\times \left[\bar{n}\left(\frac{\Delta E}{\hbar}\right) + \frac{1}{2} \pm \frac{1}{2}\right]; \qquad (2.11)$$

$\psi(\omega)$ is the frequency density function, normalized by the condition

$$\int \psi(\omega)\,d\omega + \frac{N_d}{3N} = 1,$$

where N_d is the number of localized normal vibrations of the system; the integration extends over all the quasicontinuous bands; the tilde denotes an average over all normal vibrations having the same frequency because of the strict degeneracy caused by the symmetry of the system.

If the excited states of the normal oscillators which correspond to the discrete frequencies were rigorously stationary, the function $\rho'(\Delta E, \kappa)$, which is responsible for the creation or absorption of the oscillation quantum $\hbar\omega_d$, would be given by the expression

$$\rho^1(\Delta E, \varkappa) = W\exp\left\{\pm\frac{\hbar\omega_d}{2kT}\right\}I_1\left[\sum_{(d)}\frac{R_0(\varkappa S(0; d))^2}{\hbar\omega_d}\,\text{sh}^{-1}\frac{\hbar\omega_d}{2kT}\right]$$
$$\times\left\{I_0\left[\sum_{(d)}\frac{R_0(\varkappa S(0; d))^2}{\hbar\omega_d}\,\text{sh}^{-1}\frac{\hbar\omega_d}{2kT}\right]\right\}^{-1}$$
$$\times\frac{1}{\pi}\frac{\Gamma/2}{(|\Delta E| - \hbar\omega_d)^2 + \Gamma^2/4}. \qquad (2.12)$$

At distances $\Delta E = \pm\hbar\omega_d$ from the central peak, there are satellites having the natural width and with an intensity

$$\exp\left\{\pm\frac{\hbar\omega_d}{2kT}\right\}I_1\left[\sum_{(d)}\frac{R_0(\varkappa S(0; d))^2}{\hbar\omega_d}\,\text{sh}^{-1}\frac{\hbar\omega_d}{2kT}\right]$$
$$\times\left\{I_0\left[\sum_{(d)}\frac{R_0(\varkappa S(0; d))^2}{\hbar\omega_d}\,\text{sh}^{-1}\frac{\hbar\omega_d}{2kT}\right]\right\}^{-1} \qquad (2.13)$$

times less than that of the true Mössbauer line.

When $kT/\hbar\omega_d \ll 1$, this relation, for the case of emission of a vibration quantum, becomes simply

$$(R_0/\hbar\omega_d)\sum_{(d)}(\varkappa S(0; d))^2. \qquad (2.13')$$

In real systems, the excited states of the discrete normal oscillators are not stationary even at T = 0. Because of this the levels will have a finite width, and the distribution of intensity with energy will differ from (2.12). Taking a Lorentz shape with width Γ_d for the first excited oscillator d when $kT/\hbar\omega_d \ll 1$, we have

$$\rho^1(\Delta E, \varkappa) = W\frac{R_0}{\hbar\omega_d}\sum_{(d)}(\varkappa S(0; d))^2\frac{1}{\pi}\frac{(\Gamma + \Gamma_d)/2}{(\Delta E - \hbar\omega_d)^2 + (\Gamma + \Gamma_d)^2/4},$$
$$\Delta E > 0. \qquad (2.14)$$

Usually $\hbar\omega_d \gg \Gamma_d \gg \Gamma$; then the ratio of the intensities of the satellites and the Mössbauer line will contain, in addition to (2.13'), a factor Γ/Γ_d. But the ratio of the areas under the curves corresponding to the Mössbauer peak and to the satellites will again be given by (2.13').

Now let us look at the probability of the true Mössbauer effect. Strictly speaking, the quantity W^0 of (2.9) for $T \neq 0$ actually determines a lower limit for the effect, since it does not take account of transitions in which there is a change in state of the system but no change in energy. Such transitions in the quasicontinuous spectrum give only a very small correction, of the order of the ratio Γ/Δ in the most favorable case (Δ is the width of the energy band), which was assumed to be small from the outset. The same statement applies to transitions in which quanta of the discrete and quasicontinuous spectra take part together.

The problem is somewhat more complex for transitions without a change of energy, in which only discrete normal vibrations participate. Here cases of degeneracy can occur, and consequently, when $T \neq 0$, there should be transitions with simultaneous emission and absorption of two (or more) quanta of oscillations having the same frequency but belonging to different normal vibrations. Just for this reason, the probability for the Mössbauer effect in the general case should be given by (2.10') and not by (2.9). But in most cases (for example, for isolated impurity atoms), if we exclude accidental degeneracies, the strict degeneracy due to symmetry will not be more than threefold for the localized vibrations in which a particular atom participates. The corresponding polarization vectors $S(0; d)$ can be taken to be orthogonal. Choosing one of the polarization vectors along $\mathbf{k}$ in the

case of threefold degeneracy, or along the projection of k on the plane containing the polarization vectors in the case of twofold degeneracy (in the first case the direction of the polarization vectors is arbitrary, in the second case it is arbitrary in the fixed plane), we eliminate entirely the need for including transitions between degenerate states. In this case (2.9) is a rigorous expression for the probability of the Mössbauer effect, not only when $T = 0$, but also for arbitrary T.

We note that in I we dealt precisely with the strict degeneracy of localized levels for an impurity atom in a cubic lattice. [The remark on the special choice of the directions of the polarization vectors is important only for the intermediate formulas (3.25) and (3.26) in I.]

To conclude this section, we consider some general results which can be obtained for the probability of the Mössbauer effect, irrespective of the specific form of the macroscopic system.

Let $kT \gtrsim \hbar\omega_{max}$. Then

$$2\bar{n}_\beta + 1 \approx \frac{2kT}{\hbar\omega_\beta}\left[1 + \frac{1}{12}\left(\frac{\hbar\omega_\beta}{kT}\right)^2\right]. \qquad (2.15)$$

Then

$$Z = \frac{2kTR_0}{\hbar^2}\sum_\beta\frac{(\varkappa S\,(0;\,\beta))^2}{\omega_\beta^2} + \frac{R_0}{6kT}\sum_\beta(\varkappa S\,(0;\,\beta))^2. \qquad (2.16)$$

Since the transformation (2.2) is unitary,

$$\sum_\beta S^i\,(l;\,\beta)\,S^k\,(l';\,\beta) = \delta^{ik}\delta_{ll'}, \qquad (2.17)$$

while the last term in (2.16) is equal simply to $R_0/6kT$.

We multiply both sides of (2.3) by the reciprocal matrix $[\bar{A}^{-1}(m, l)]^{ji}$ and sum over l and i:

$$S^l\,(m;\,\beta)/\omega_\beta^2 = \sum_l[\bar{A}^{-1}\,(m,\,l)]^{li}\,S^i\,(l;\,\beta).$$

Set $m = 0$. Multiply both sides of the equation by $S^k(0;\,\beta)$ and sum over β. Interchanging the order of the summations on the right, and using (2.17), we finally get

$$\sum_\beta\omega_\beta^{-2}S^l(0;\,\beta)\,S^k(0;\,\beta) = [\bar{A}^{-1}(0,\,0)]^{lk} = m_0\,[A^{-1}(0,\,0)]^{lk}. \qquad (2.18)$$

Substituting these expressions in (2.16), we arrive at the expression

$$Z = 2kTR_0m_0\hbar^{-2}\,[A^{-1}(0;\,0)]^{lk}\,\varkappa^l\varkappa^k + R_0/6kT. \qquad (2.19)$$

A very important conclusion follows from (2.19) —in the absence of discrete frequencies, the probability for the Mössbauer effect when $kT > \hbar\omega_{max}$ is independent of the mass of the radiating (or absorbing) nucleus, and is determined only by the interaction matrix. Only a correction term, which is

already small when $kT = \hbar\omega_{max}$, is dependent on the value of m_0 (and only on m_0).

In particular, if an impurity atom of mass m_j replaces an atom of mass m_0 at the j-th site of an arbitrary unit cell of the regular lattice, then if the force constants are unchanged

$$Z = \frac{2kTR_0m_0}{\hbar^2m_j}\,\frac{v_0}{(2\pi)^3}\sum_\alpha\int d^3f\,\frac{|\varkappa V_j\,(\mathbf{f},\,\alpha)\,|^2}{\omega^2\,(\mathbf{f},\,\alpha)} + \frac{R_0}{6kT}, \qquad (2.20)$$

where $V_j(\mathbf{f},\,\alpha)$ are complex polarization vectors. The first term in (2.20) is precisely the usual expression for the Mössbauer effect on the j-th atom of the regular lattice at high temperatures (cf. [5] or [6]) and is completely independent of the mass of the impurity nucleus.

When discrete frequencies are present, W in (2.10′) in the classical limit ($kT \gtrsim \hbar\omega_{max}$) generally remains dependent on m_0 through the argument of I_0, which in this case has the form

$$\frac{2kTR_0}{(\hbar\omega_d)^2}\sum_{(d)}(\varkappa S\,(0;\,d))^2\left[1 - \frac{1}{24}\left(\frac{\hbar\omega_d}{kT}\right)^2\right]. \qquad (2.21)$$

If the quantity (2.21) is small compared to unity for a reasonable range of temperatures and for all discrete frequencies, $W \approx e^{-Z}$, just as in the case where the discrete frequencies are absent.

In the opposite limiting case, when $kT > \hbar\omega_d$

$$\frac{2kTR_0}{(\hbar\omega_d)^2}\sum_{(d)}\,(\varkappa S\,(0;\,d))^2 \gg 1, \qquad (2.22)$$

using the asymptotic form for I_0, we get the following expression:

$$W_2 = \prod_d{}'\left[\frac{4\pi kTR_0}{(\hbar\omega_d)^2}\sum_{(d)}(\varkappa S\,(0;\,d))^2\right]^{-1/2}. \qquad (2.23)$$

The result in (2.23) can radically change the temperature dependence of the Mössbauer effect: when condition (2.22) is satisfied, the falloff of W_2 with temperature when $kT > \hbar\omega_d$ will not be exponential, but rather will follow a power law. The dependence will be weakest when there is only one discrete frequency (degenerate or nondegenerate) and will be $\sim 1/\sqrt{T}$.

3. ISOLATED IMPURITY NUCLEUS IN A REGULAR LATTICE

Now let us consider a regular monatomic lattice with arbitrary isolated impurity atoms.

In I we found the probability for resonance absorption (emission) of a γ quantum by an impurity atom, without a change in state of the crystal, for an arbitrary ratio of the mass m' of the impurity atom to the mass m of the atoms of the host lattice, and used a simple one-parameter model for the

change in the force constants. The model used in I for the change in force constants is very convenient for formal analysis, and allows one to obtain results over a wide range of variation of the force constants determined by the requirement that the potential energy be positive definite [for a cubic crystal this interval is $(\langle \omega_0^2 \rangle \langle \omega_0^{-2} \rangle - 1)^{-1} < \gamma < 1]$. But this model is too artificial. Among the solutions of Eq. (3.5) of I, there is no constant vector corresponding to the zero eigenfrequency which occurs when the individual atoms are fixed absolutely in space. In comparing with experimental data for arbitrary m'/m, it is therefore necessary to use the results of I only for $\gamma = 0$, since then the theory is free of the defect which was just mentioned.

In the most general case, $A^{ik}(\mathbf{r_n}, \mathbf{r_{n'}})$ can be written in the following form:

$$A^{ik}(\mathbf{r_n}, \mathbf{r_{n'}}) = A_0^{ik}(\mathbf{r_n} - \mathbf{r_{n'}}) + \gamma \Lambda^{ik}(\mathbf{r_n}, \mathbf{r_{n'}}), \quad (3.1)$$

where $\gamma \Lambda^{ik}(\mathbf{r_n}, \mathbf{r_{n'}})$ is a perturbation operator, which satisfies the usual unitarity relations and is damped rapidly with increasing $|\mathbf{r_n}|$ and $|\mathbf{r_{n'}}|$ (the impurity atom is at the coordinate origin. The equation for the vibrations is

$$m(1 - \varepsilon \delta_{n0}) \omega_\beta^2 w_\mathbf{n}^i(\beta)$$

$$= \sum_{n'} A_0^{ik}(\mathbf{r_n} - \mathbf{r_{n'}}) w_{n'}^k(\beta) + \gamma \sum_{n'} \Lambda^{ik}(\mathbf{r_n}, \mathbf{r_{n'}}) w_{n'}^k(\beta).$$
$$(3.2)$$

In order to be able to visualize the final results, we restrict ourselves to the case where the ratio m'/m is arbitrary but the change in the force constants is small, and find the solution to terms linear in γ.

Let us write $S^i(\mathbf{n}; \beta) = \sqrt{m_\mathbf{n}}\, w_\mathbf{n}^i(\beta)$ in the following form:

$$S^i(\mathbf{n}; \beta) = S_0^i(\mathbf{n}; \beta) + \gamma S_1^i(\mathbf{n}; \beta). \quad (3.3)$$

Here $S_0(\mathbf{n}; \beta)$ is a normalized solution of (3.2) for $\gamma = 0$. In accordance with the results of the preceding section, we shall be interested only in quantities of the form $S^i(0; \beta) S^k(0; \beta)$, which, in the linear approximation in γ, are expressed as

$$S^i(0; \beta) S^k(0; \beta) = S_0^i(0; \beta) S_0^k(0; \beta)$$

$$+ \gamma [S_0^i(0; \beta) S_1^k(0; \beta) + S_0^k(0; \beta) S_1^i(0; \beta)]. \quad (3.4)$$

From the form of (3.4) it follows that for our purposes it is sufficient to consider only those normal vibrations β in which the displacement of the impurity atom is different from zero when $\gamma = 0$.

In the regular lattice, each eigenvalue $\omega_0^2(\mathbf{f}, \alpha)$ is degenerate, and the multiplicity θ is determined by the number of vectors $\mathbf{f}$ which are connected with one another by the transformations of the point symmetry group of the crystal (the star of the irreducible representation). Let us denote the set of values $\omega_0^2(\mathbf{f}, \alpha)$ belonging to the same star and some α, by ω_{0p}^2, where the subscript p labels the frequencies in increasing order.

For simplicity, let us restrict our treatment to crystals with symmetry not lower than rhombic. Then in the presence of the impurity atom ($\gamma = 0$) three frequencies split off from each level p, and only for these frequencies is $S^i(0; \beta) \neq 0$. In place of the index β, we shall use the double index τ, p, where τ runs through the values 1, 2, 3. In accordance with the results found in I, we get

$$S_0^i(\mathbf{n}; \tau, p) = L_{\tau p}^{1/2} j_\tau^k \frac{1}{N} \sum_{f, \alpha} \left[\varepsilon \omega_{\tau p}^2 \frac{e^i(\mathbf{f}, \alpha) e^k(\mathbf{f}, \alpha)}{\omega_{\tau p}^2 - \omega_0^2(\mathbf{f}, \alpha)} \right.$$

$$\left. + \frac{1}{3}(\sqrt{1 - \varepsilon} - 1) \delta^{ik} \right] \cos \mathbf{f r_n}, \quad (3.5)$$

where j_τ are unit vectors along the principal axes of symmetry of the crystal, while $L_{\tau p} \equiv \partial \ln \omega_{\tau p}^2 / \partial \varepsilon$ is determined from the solution of the equation

$$1 = \frac{\varepsilon \omega_{\tau p}^2}{N} \sum_{f, \alpha} \frac{e^\tau(\mathbf{f}, \alpha) e^\tau(\mathbf{f}, \alpha)}{\omega_{\tau p}^2 - \omega_0^2(\mathbf{f}, \alpha)} \quad (3.6)$$

(no summation over τ). From (3.2), using standard perturbation theory, we find for $S_1^i(0; \tau, p)$

$$S_1^i(0; \tau, p) = \sum_{\tau' p'}' \frac{\Lambda_{\tau p, \tau' p'}}{\omega_{\tau p}^2 - \omega_{\tau' p'}^2} S_0^i(0; \tau', p'), \quad (3.7)$$

where

$$\Lambda_{\tau p, \tau' p'} = \sum_{n, n'} S_0^i(\mathbf{n}; \tau, p)(m_\mathbf{n} m_{n'})^{-1/2} \Lambda^{ik}(\mathbf{r_n}, \mathbf{r_{n'}}) S_0^k(\mathbf{n'}; \tau', p'). \quad (3.8)$$

Let us consider one-phonon excitations. For the quasicontinuous spectrum, according to (2.10) and using (3.5) and (3.6), we have

$$p^1(\Delta E, \varkappa)/W = p_0^1(\Delta E, \varkappa)/W + \gamma p_1^1(\Delta E, \varkappa)/W; \quad (3.9)$$

$$p_0^1(\Delta E, \varkappa)/W = 2R'(1 - \varepsilon) \hbar^{-2} \sum_\tau (\varkappa j_\tau)^2 g(\omega^2)$$

$$\times f_\tau(\omega^2) [\bar{n}(\omega) + \frac{1}{2} \pm \frac{1}{2}]|_{\omega = |\Delta E|/\hbar}, \quad (3.10)$$

$$p_1^1(\Delta E, \varkappa)/W = 4\bar{R}'(1 - \varepsilon) \hbar^{-2} g(\omega^2) \sum_{\tau, \tau'}' (\varkappa j_\tau)(\varkappa j_{\tau'}) \sqrt{f_\tau(\omega^2)}$$

$$\times \left[\int_0^{\omega_{0\,max}^2} d\omega'^2 \frac{g(\omega'^2) \sqrt{f_{\tau'}(\omega'^2)} \Lambda_{\tau, \tau'}(\omega^2, \omega'^2)}{\omega^2 - \omega'^2} + \frac{\Lambda_{\tau\tau'}(\omega^2, \omega_{D\tau'}^2)}{\omega^2 - \omega_{D\tau'}^2} L_{D\tau'}^{1/2} \right]$$

$$\times [\bar{n}(\omega) + \frac{1}{2} \pm \frac{1}{2}]|_{\omega = |\Delta E|/\hbar}. \quad (3.11)$$

Here $g(\omega^2)$ is the distribution function for the squares of the frequences of the regular lattice;

$$R' \equiv R_0 = \hbar^2 k^2 / 2m'; \quad (3.12)$$

$f_\tau(\omega^2)$

$$= \frac{g_\tau(\omega^2)}{g(\omega^2)} \left\{ \left[1 - \varepsilon\omega^2 \int_0^{\omega^2_{0max}} \frac{g_\tau(\omega_0^2)\,d\omega_0^2}{\omega^2 - \omega_0^2} \right]^2 + [\pi\varepsilon\omega^2 g_\tau(\omega^2)]^2 \right\}^{-1},$$

$$(3.13)$$

where

$$\Lambda_{\tau,\tau'}(\omega^2, \omega'^2) = \lim_{N\to\infty} 3N\vartheta^{-1}\Lambda_{\tau p, \tau'p'},$$

$$g_\tau(\omega^2) = \frac{v_0}{(2\pi)^3} \sum_\alpha \int_{\omega_0^2(f,\alpha)=\omega^2} d\Omega_\omega \frac{[e^\tau(f,\alpha)]^2}{|\nabla\omega_0^2(f,\alpha)|}. \qquad (3.13')$$

We note that from now on, for cubic lattices, we shall choose the set of vectors j_τ mentioned above, where one of them is along κ. This automatically eliminates the problem of transitions between levels belonging to the same p. Similarly, in the case of uniaxial crystals we should choose one of the polarization vectors in the plane perpendicular to the symmetry axis to be along the projection of the vector κ onto this plane.

Now let us treat ρ' for the discrete part of the spectrum, for which, in accordance with (2.13) and (2.14), we determine the value of $S^i(0; d) S^k(0; d)$. We shall restrict ourselves to low temperatures, for which the condition $kT/\hbar\omega_d \ll 1$ is satisfied. Then for each level $d = \tau$ (it assumed that in the absence of degeneracy the level spacing is larger than their width) we have ($\Delta E > 0$)

$$\rho_0^1(\Delta E, \varkappa)/W = R'(1 - \varepsilon)(\varkappa j_\tau)^2 (\hbar\omega_{D\tau})^{-1} L_{D\tau}\frac{1}{\pi}$$

$$\times \frac{(\Gamma + \Gamma_{D\tau})/2}{(\Delta E - \hbar\omega_{D\tau})^2 + (\Gamma + \Gamma_{D\tau})^2/4}, \qquad (3.14)$$

where we have introduced the notation $L_{D\tau} \equiv \partial\ln\omega_{D\tau}^2/\partial\epsilon$. We give the value of $\rho_1^1(\Delta E, \kappa)$ corresponding to the case of cubic symmetry of the host:

$$\rho_1^1(\Delta E, \varkappa)/W$$

$$= 2R'(1 - \varepsilon)\frac{1}{\hbar\omega_D} L_D^{1/2} \int_0^{\omega_{0max}^2} d\omega^2 \frac{g(\omega^2)\sqrt{f(\omega^2)}\Lambda(\omega_D^2, \omega^2)}{\omega_D^2 - \omega^2}$$

$$\times \frac{1}{\pi} \frac{(\Gamma + \Gamma_D)/2}{(\Delta E - \hbar\omega_D)^2 + (\Gamma + \Gamma_D)^2/4},$$

$$\Delta E > 0; \qquad \Lambda(\omega_D^2, \omega^2) = \lim_{N\to\infty} (3N\vartheta^{-1})^{1/2}\Lambda_{D\tau, \tau p}\big|_{\omega_p^2=\omega^2},$$

$$j_\tau = \varkappa, \quad L_D \equiv \partial\ln\omega_D^2/\partial\varepsilon. \qquad (3.14')$$

Now we determine the probability W^0 (2.9) for the Mössbauer effect. Using (3.4), we write Z in the form

$$Z = R' \sum_{\tau p} (\hbar\omega_{\tau p})^{-1} (\varkappa S_0(0; \tau, p))^2 [2\bar{n}(\omega_{\tau p}) + 1]$$

$$+ 2\gamma R' \sum_{\tau p} (\hbar\omega_{\tau p})^{-1} (\varkappa S_0(0; \tau, p))(\varkappa S_1(0; \tau,p))[2\bar{n}(\omega_{\tau p})$$

$$+ 1] = Z_0 + \gamma Z_i.$$

For $\gamma = 0$,

$$W^0 = \exp\left\{ -R'(1 - \varepsilon)\sum_{\tau p}(\varkappa j_\tau)^2\frac{1}{\hbar\omega_{\tau p}}L_{\tau p}[2\bar{n}(\omega_{\tau p}) + 1] \right\}$$

$$\times \prod_{\tau=1}^3 I_0\left[R'(1 - \varepsilon)(\varkappa j_\tau)^2\frac{1}{\hbar\omega_{D\tau}}L_{D\tau}\,\mathrm{sh}^{-1}\frac{\hbar\omega_{D\tau}}{2kT} \right], \qquad (3.15)$$

where $\sum_{\tau p}$, includes a summation over the discrete frequencies.

After changing from summation over the quasi-continuous spectrum to an integration, we have

$$W^0 = e^{-Z}\prod_{\tau=1}^3 I_0\left[R'(1 - \varepsilon)(\varkappa j_\tau)^2 (\hbar\omega_{D\tau})^{-1}L_{D\tau}\,\mathrm{sh}^{-1}\frac{\hbar\omega_{D\tau}}{2kT} \right],$$

$$Z = Z' + Z'',$$

$$Z' = R'(1 - \varepsilon)\sum_\tau(\varkappa j_\tau)^2$$

$$\times \int_0^{\omega_{0max}^2} (\hbar\omega)^{-1} g(\omega^2) f_\tau(\omega^2)(2\bar{n}(\omega) + 1)\,d\omega^2,$$

$$Z'' = R'(1 - \varepsilon)\sum_\tau(\varkappa j_\tau)^2 (\hbar\omega_{D\tau})^{-1}L_{D\tau}(2\bar{n}(\omega_{D\tau}) + 1). \quad (3.15')$$

These expressions are the same as those found in I. For $\gamma \neq 0$, we give only the value of Z_1 corresponding to $kT > \hbar\omega_{0\,max}$:

$$Z_1 = -2kTR'(1 - \varepsilon)\hbar^{-2}\varkappa^i\varkappa^k N^{-2}\sum_{f,\alpha}\sum_{f',\alpha'}\frac{e^i(f, \alpha)\Lambda_{f\alpha, f'\alpha'}\,e^k(f', \alpha')}{\omega_0^2(f, \alpha)\omega_0^2(f', \alpha')},$$

$$(3.16)$$

where

$$\Lambda_{f\alpha, f'\alpha'} = m^{-1}\sum_n\sum_{n'}\Lambda^{ls}(r_n, r_{n'})\,e^l(f, \alpha)\,\epsilon^s(f', \alpha')$$

$$\times \exp\{-if r_n + if' r_{n'}\}. \qquad (3.17)$$

4. DISCUSSION OF RESULTS

Let us examine the results obtained in the preceding sections and in I. For simplicity we consider only the case of $\gamma = 0$.

All the formulas given depend essentially on the quantity

$$L_{\tau p} \equiv \partial\ln\omega_{\tau p}^2/\partial\varepsilon.$$

It appears that one can give a simple normalization condition for this quantity. In the complete

system of eigenfunctions for the vibrational problem with an impurity atom, the functions (3.5) differ from zero only for $n = 0$. If we make use of the completeness relation,

$$\sum_{\beta} S^l (0; \beta) S^k (0; \beta) = (1 - \varepsilon) \sum_{\tau p} L_{\tau p} j_\tau^l j_\tau^k = \delta^{ik}.$$

Using the fact that in crystals with higher symmetry than rhombic j_τ can be chosen along the principal axes of symmetry of the crystal independently of p, we find

$$(1 - \varepsilon) \sum_p L_{\tau p} = 1, \qquad \tau = 1, 2, 3. \qquad (4.1)$$

To each τ there corresponds no more than one discrete frequency. Thus in the general case, using (3.13) and changing from summation to integration over the quasicontinuous spectrum, we have

$$(1 - \varepsilon) \int_0^{\omega_0^2 \, max} d\omega^2 g (\omega^2) f_\tau (\omega^2) + (1 - \varepsilon) L_{D\tau} = 1. \qquad (4.2)$$

In the cubic case, the three equations are identical ($f_\tau = f$).

We note that it follows from (4.2) that one can determine $L_{D\tau}$ not from the dependence of $\omega_{D\tau}^2$ on ε, but from an integral over f_τ.

Let us consider the probability for the Mössbauer effect. For $T = 0$ it follows from (3.15) that W^0 depends on the average of the quantity $1/\omega$ over the quasicontinuous and discrete spectra, where the statistical weight is the quantity $(j_\tau \cdot \kappa)^2 \times (1 - \varepsilon) L_{\tau p}$.

Consider the case of a light radiator. As the ratio m'/m decreases (starting from some critical value $\varepsilon_c > 0$, at which the discrete frequency $\omega_{D\tau}$ appears), there is a continuous rise of $(1 - \varepsilon) L_{D\tau}$, and at the same time a decrease of the total amplitude of oscillation of the impurity atom in the quasicontinuous spectrum. One can show, for example, that in a cubic crystal, to terms of order $(1 - \varepsilon)^2$, the relation

$$(1 - \varepsilon) \int_0^{\omega_0^2 \, max} d\omega^2 g (\omega^2) f (\omega^2) = (1 - \varepsilon) \frac{\langle \omega_0^4 \rangle - \langle \omega_0^2 \rangle^2}{\langle \omega_0^2 \rangle^2} \qquad (4.2')$$

holds. Thus with increasing ε, the quantity $(1 - \varepsilon) L_{D\tau}$ approaches unity, while the impurity atom begins to vibrate mainly at the discrete frequencies. Since the frequency density function is localized in the region of high discrete frequencies, this is a most favorable situation for obtaining a minimum (for given R') value of Z and at the same time a maximum probability for the Mössbauer effect.

Noting that for small $1 - \varepsilon$,

$$\omega_{D\tau}^2 = \frac{3 \langle \omega_0^2 e^\tau e^\tau \rangle}{1 - \varepsilon} \left[1 + (1 - \varepsilon) \frac{\langle \omega_0^4 e^\tau e^\tau \rangle - 3 \langle \omega_0^2 e^\tau e^\tau \rangle^2}{3 \langle \omega_0^2 e^\tau e^\tau \rangle^2} \right],$$

$$\langle \omega_0^{2n} e^\tau e^\tau \rangle \equiv (3N)^{-1} \sum_{f, \alpha} \omega_0^{2n} (f, \alpha) e^\tau (f, \alpha) e^\tau (f, \alpha), \qquad (4.3)$$

we have for W_2^0

$$W_2^0 = \exp \left\{ - \frac{R' (1 - \varepsilon)^{1/2}}{\hbar} \sum_\tau \frac{(j_\tau \kappa)^2}{\sqrt{3} \langle \omega_0^2 e^\tau e^\tau \rangle^{1/2}} \right.$$

$$\left. \times \left[1 - \frac{3}{2} (1 - \varepsilon) \frac{\langle \omega_0^4 e^\tau e^\tau \rangle - 3 \langle \omega_0^2 e^\tau e^\tau \rangle^2}{3 \langle \omega_0^2 e^\tau e^\tau \rangle^2} \right] \right\}. \qquad (4.4)$$

In general the effect is definitely anisotropic; from the value of W^0 in a single crystal for three values of κ which do not lie in a plane, one can get the values of $\omega_{D\tau}$.

For a cubic crystal, (4.4) simplifies and reduces to

$$W_2^0 = \exp \left\{ - \frac{R' (1 - \varepsilon)^{1/2}}{\hbar \langle \omega_0^2 \rangle^{1/2}} \left[1 - \frac{3}{2} (1 - \varepsilon) \frac{\langle \omega_0^4 \rangle - \langle \omega_0^2 \rangle^2}{\langle \omega_0^2 \rangle^2} \right] \right\}, \qquad (4.4')$$

which agrees with I.

The appearance in the exponent of the small factor $(1 - \varepsilon)^{1/2} = (m'/m)^{1/2}$ is very important for obtaining an observable Mössbauer effect on relatively light impurity nuclei. The value in (4.4) is a limit, and W_2^0 can increase for given R' only if the force constants change. We note that $\langle 1/\omega_0 \rangle > \langle \omega_0^2 \rangle^{-1/2}$ and thus, compared to the regular lattice, there is an additional increase in the effect.

Now let us consider an inelastic transition with excitation of a discrete frequency. To each frequency $\omega_{D\tau}$ there will correspond the appearance in the resonance absorption (emission) intensity curve of a sharp peak, displaced from the Mössbauer peak by $\Delta E = \hbar \omega_{D\tau}$. According to (2.13), the ratio of the areas under these peaks will be

$$R' (1 - \varepsilon) (\hbar \omega_{D\tau})^{-1} (j_\tau \kappa)^2 L_{D\tau}. \qquad (4.5)$$

Consequently, for sufficiently small m'/m, this ratio will be simply $R' (\hbar \omega_{D\tau})^{-1} (j_\tau \cdot \kappa)^2$, and in a cubic crystal, $R'/\hbar \omega_D$. But this quantity, though in most cases less than unity, can be comparable to unity. Thus the integral intensity in each satellite may be comparable with the corresponding quantity for the Mössbauer line.

For the experimental verification of such a transition, the differential intensity given by (3.14) is important. It is easy to see that when the integral intensity is large, the value of the width $\Gamma_{D\tau}$ will be the decisive factor.

We have made a calculation of the width of the excited level of a localized normal oscillator in crystals which results from anharmonicity.[7] We treated crystals with cubic symmetry and included nearest neighbor interactions. We found that even under favorable energetic conditions, the width for the decay of a quantum of the localized vibration into two excitations of the quasicontinuous spectrum was 10^{-2} of $\hbar\omega_D$ when $kT \ll \hbar\omega_{0\,max}$. For ω_D close to $2\omega_{0\,max}$, and all the more for $\omega_D > 2\omega_{0\,max}$, the value of Γ_D due to anharmonicity is even lower. Thus, at low temperatures the differential intensity corresponding to an inelastic transition with excitation of a discrete frequency, which depends on the ratio Γ/Γ_D, will apparently be at least two orders of magnitude stronger than the intensity for one-phonon transitions in the quasicontinuous spectrum, which is determined by $\Gamma/\hbar\omega_{0\,max}$. We note that if the source and absorber are identical, under certain circumstances one can increase the effect significantly by making the peaks for one-phonon emission and absorption coincide.

When $m'/m > 1 - \epsilon_c$, there are no discrete frequencies, and the second term on the left of (4.2) vanishes. As m'/m increases there is a continual redistribution of $f_\tau(\omega^2)$ into the region of lower frequencies. The picture becomes particularly clear for $m'/m \gg 1$. Then f_τ becomes a δ function localized in the low-frequency region.

As an example, let us consider the case of cubic symmetry, and take for f the corresponding value from (3.13) ($g_\tau = g$). It is easy to see that when $|\epsilon| \gg 1$ ($\epsilon < 0$), the denominator in (3.13) has a sharp, narrow minimum at $\omega_0^2/\omega_{0\,max}^2 \sim 1/|\epsilon|$. Using the fact that in this range $g(\omega^2) = d\sqrt{\omega^2}/\omega_{0\,max}^3$, after making some transformations we arrive at the following approximate expression:

$$f(x) \approx \frac{\langle x^{-1}\rangle^{1/2}}{\pi d\,|\epsilon|^{1/2}} \frac{\lambda}{(x - x_0)^2 + \lambda^2}, \quad x = \frac{\omega_0^2}{\omega_{0\,max}^2}, \quad (4.6)$$

where

$$x_0 = 1/|\epsilon|\langle x^{-1}\rangle, \quad \lambda = \pi d/|\epsilon|^{1/2}\langle x^{-1}\rangle^{5/2}. \quad (4.7)$$

Thus the impurity atom oscillates primarily in a comparatively narrow range of frequencies, the center of this region on the ω^2 scale being displaced toward lower frequencies by an amount proportional to $|\epsilon|^{-1}$. Such a localization of f obviously leads to a reduction in the probability for the Mössbauer effect as compared to that for a regular crystal with the frequency spectrum of the host but with atoms of mass m'.

The limiting value of W^0 in a cubic crystal, which is easily found using (4.6) [cf. (5.17) in I],

$$W^0 = \exp\left\{-\frac{R'}{\hbar}\left\langle\frac{1}{\omega_0^2}\right\rangle^{1/2}|\epsilon|^{1/2}\right\}, \quad (4.8)$$

clearly shows this effect. On the other hand, it follows from (4.8) that as the mass of the radiating atom increases while the mass of the lattice atoms is kept fixed, Z drops continuously, though only as $1/\sqrt{m'}$.

We note that the probability for the Mössbauer effect in the regular lattice is related to the moment $\langle 1/\omega_0\rangle$, the limiting value for $m'/m \ll 1$ is determined by $\langle\omega_0^2\rangle$ while the limit for $m'/m \gg 1$ is determined by $\langle\omega_0^{-2}\rangle$. Thus in these three cases the value of W^0 gives three different moments of the spectrum of the ideal lattice.

Now let us consider one-phonon transitions in the quasicontinuous spectrum. According to (3.9'), the determination of $\rho'(\Delta E, \kappa)$ for κ along the principal axes of the crystal enables us to determine

$$\varphi_\tau(\omega^2) = g(\omega^2)\,f_\tau(\omega^2) \quad (4.9)$$

as a function of frequency.

Knowledge of $\varphi_T(\omega^2)$ enables a complete determination of the nature of the vibrations of the impurity atom.

But the question of greatest interest is the possibility of determining from φ_T the frequency distribution function $g(\omega^2)$ for the ideal lattice. Consider the case of cubic symmetry. From (3.13) it follows that f is related to the parameters of the host only through $g(\omega^2)$. Thus there is a functional relation between φ and g, and consequently there is a possibility of determining $g(\omega^2)$ from the results of measurements of one-phonon processes in the resonance absorption (emission) of γ quanta by impurity nuclei. [In the anisotropic case it is necessary to make measurements for three values of κ[5] and to determine each $g_T(\omega^2)$ independently.]

It is interesting to note that, in accordance with (4.6), when $m'/m \gg 1$ the probability for one-phonon excitations has a marked resonance character for $\omega = \sqrt{x_0}\,\omega_{0\,max}$, which is very attractive for experimental observation.

We note that when the force constants change the problem of reconstructing the frequency distribution function is much more complicated. But if there is a considerable difference between m' and m, one can in most cases neglect the change in the force constants. The comparison with experiment in the next Section is an indirect confirmation of this. We should especially state that all the complications which enter in the problem of experimentally determining $g(\omega^2)$ for the case of regular crystals[5,8] are completely applicable for the case where im-

purities are present. We shall not spend time on this problem.

Now let us go on to the treatment of the temperature dependence. Let $kT > \hbar\omega_{DT}$ (in the absence of discrete frequencies $kT > \hbar\omega_{0\,max}$) so that (2.15) is valid. One can prove rigorously that for any vibration frequency spectrum the relation

$$\sum_p \frac{1}{\omega_{\tau p}^2} L_{\tau p} + \frac{1}{\omega_{D\tau}^2} L_{D\tau} = \frac{1}{N}\sum_{f,\alpha} \frac{e^\tau(f,\,\alpha)\,e^\tau(f,\,\alpha)}{\omega_0^2(f,\,\alpha)} \qquad (4.10)$$

is valid.

We then have from (3.15)

$$W^0 = \exp\left\{ -2kTR'(1-\varepsilon)\hbar^{-2}N^{-1}\sum_{f,\alpha}\omega_0^{-2}(f,\,\alpha) \right.$$

$$\times(\varkappa e(f,\,\alpha))^2 - R'/6kT\right\}\prod_{\tau=1}^3 I_0\left[\frac{2kTR'(1-\varepsilon)(\varkappa j_\tau)^2}{(\hbar\omega_{D\tau})^2}L_{D\tau}\right]$$

$$(4.11)$$

in complete agreement with (2.19) and (2.20).

For a host with cubic symmetry

$$W^0 = \exp\left\{ -\frac{2kTR'(1-\varepsilon)}{\hbar^2}\left\langle\frac{1}{\omega_0^2}\right\rangle - \frac{R'}{6kT}\right\}I_0$$

$$\times\left[\frac{2kTR'(1-\varepsilon)}{(\hbar\omega_D)^2}L_D\right]. \qquad (4.11')$$

We note again the complete disappearance of the dependence on mass of the radiating nucleus for the main term in (4.11) and (4.11') when there are no discrete frequencies $[R'(1-\varepsilon) = \hbar^2k^2/2m]$.

For a heavy radiator (no discrete frequencies) the classical limit (4.11) is reached at much lower temperatures. In fact when $|\varepsilon| \gg 1$ the impurity atom vibrates in a narrow range of frequencies around $\omega = \omega_{0\,max}/|\varepsilon|^{1/2}\langle x^{-1}\rangle^{-1/2}$ [cf. (4.7)]. Consequently it is sufficient to have

$$kT > \hbar\omega_{0\,max}/|\varepsilon|^{1/2}\langle x^{-1}\rangle^{1/2} \qquad (4.12)$$

(since $\langle x^{-1}\rangle > 1$). Thus for a heavy radiator the drop in the effect with temperature occurs much more abruptly than for an ideal lattice.

We note that in a cubic crystal when $m'/m \gg 1$, because f has the form (4.6), we can immediately obtain an approximate expression for W^0 which is valid for any temperature:

$$W^0 = \exp\left\{ -\frac{R'}{\hbar}\left\langle\frac{1}{\omega_0^2}\right\rangle^{1/2}|\varepsilon|^{1/2}(2\bar n_0 + 1)\right\}, \qquad (4.13)$$

where $\bar n_0$ is determined for $\omega_0 = \sqrt{x_0}\,\omega_{0\,max}$.

If discrete frequencies appear, so that the impurity atom vibrates mainly at these discrete frequencies, the temperature dependence is sharply reduced. With increasing temperature the factors in the form of Bessel functions in (4.11) begin to increase.

When $m'/m \ll 1$, $(1-\varepsilon)L_{DT} \approx 1$, and the argument of I_0 is determined by the ratio $2kTR'/(\hbar\omega_{DT})^2$. If this ratio is small because $R'/\hbar\omega_{DT} \ll 1$, I_0 can be replaced by unity. But if $2kTR'/(\hbar\omega_{DT})^2 \gg 1$,

$$W^0 = e^{-Z'}\prod_{\tau=1}^3\left[\frac{4\pi kTR'(1-\varepsilon)(\varkappa j_\tau)^2}{(\hbar\omega_{D\tau})^2}L_{D\tau}\right]^{-1/2}. \qquad (4.14)$$

Thus in place of the exponential, W_2^0 gives a factor with a $1/T^{3/2}$ dependence. In a uniaxial crystal (3.15) will contain the product of only two Bessel functions, and the factor multiplying $e^{-Z'}$ in (4.14) will be $\sim 1/T$. Finally, for a cubic crystal only one Bessel function remains, and the factor in (4.14) is $\sim 1/T^{1/2}$. In this last case, using (4.10) [cf. (4.11')], we have

$$W^0 = \exp\left\{ -\frac{2kTR'}{\hbar^2}\left(\left\langle\frac{1}{\omega_0^2}\right\rangle - \frac{1}{\omega_D^2}L_D\right)\right.$$

$$\left. -\frac{R'}{6kT}\left[1 + \frac{1}{2}(1-\varepsilon)L_D\right]\right\}\left[\frac{4\pi kTR'(1-\varepsilon)}{(\hbar\omega_D)^2}L_D\right]^{-1/2}.$$

$$(4.15)$$

Using (4.3) one can easily show that for small $1-\varepsilon$ the relation

$$L_D/\omega_D^2 = \frac{1}{\langle\omega_0^2\rangle}\left[1 - 2(1-\varepsilon)\frac{\langle\omega_0^4\rangle - \langle\omega_0^2\rangle^2}{\langle\omega_0^2\rangle^2}\right] \qquad (4.16)$$

is valid.

5. LATTICE WITH NEAREST NEIGHBOR INTERACTIONS BETWEEN ATOMS. COMPARISON WITH EXPERIMENT.

Let us consider a crystal with a simple unit cell, including nearest neighbor interactions of both the central and noncentral type. Such a model has been used many times for the general analysis of vibrations of monatomic and diatomic cubic lattices, [9] and also for determining the probability of the Mössbauer effect. [10] This model was generalized in [11] to the case of rhombic and tetragonal symmetry. If one uses the results of this work, for all three vibration branches, whose polarization directions are fixed and coincide with the principal axes, we have expressions for the dispersion law and the partial frequency distribution function (3.13')

$$\omega_0^2(f,\,\alpha) = \frac{2}{m}\sum_{\beta=1}^3\lambda_{\alpha\beta}(1 - \cos\varphi_\beta), \qquad \varphi_\beta = fa_\beta,$$

$$g_\alpha(x) = \frac{2}{\pi}\int_0^\infty\cos[(2x-1)p]\prod_{\beta=1}^3 J_0\left(\lambda_{\alpha\beta}\left[\sum_{\gamma=1}^3\lambda_{\alpha\gamma}\right]^{-1}p\right)dp.$$

$$(5.1)$$

Here

$$x = \omega_{0\alpha}^2/\omega_{0\alpha\,max}^2, \qquad \omega_{0\alpha\,max}^2 = \frac{4}{m}\sum_{\beta=1}^3\lambda_{\alpha\beta}.$$

The coefficients $\lambda_{\alpha\beta}$ are related to the matrix elements $A_0^{ik}(\mathbf{r_n})$ as follows:

$$A_0^{ik}(\mathbf{a}_s) = A_0^{ik}(-\mathbf{a}_s) = -\lambda_{is}\delta^{ik},$$

where $\mathbf{a}_s$ (s = 1, 2, 3) denote the lattice basis vectors. All other matrix elements $A_0^{ik}(\mathbf{r_n})$, except

$$A_0^{ik}(0) = -2\sum_{s=1}^{3} A_0^{ik}(\mathbf{a}_s),$$

are equal to zero. The function $g_\alpha(x)$ is normalized by the condition

$$\int_0^1 g_\alpha(x)\,dx = 1.$$

The probability of the Mössbauer effect for an isolated impurity atom in a crystal will be determined by (3.15′) with $\tau \equiv \alpha$. The discrete frequencies are determined from three independent equations

$$x\varepsilon \int_0^1 \frac{g_\alpha(x')\,dx'}{x-x'} = 1, \qquad x > 1, \qquad (5.2)$$

after which we immediately find $\partial \ln x_{D\alpha}/\partial\epsilon$:

$$\frac{\partial \ln x_{D\alpha}}{\partial\varepsilon} = \frac{1}{\varepsilon}\left[x_{D\alpha}\varepsilon\int_0^1 \frac{g_\alpha(x)\,dx}{(x_{D\alpha}-x)^2} - 1\right]^{-1}. \qquad (5.3)$$

Using (5.1)–(5.3), we have made numerical computations of the probability for the Mössbauer effect in the case of cubic symmetry, assuming that all the constants for the central interaction are the same, β_1, and all those for the noncentral interaction are the same, β_2 (i.e., $\beta_1 = \lambda_{11} = \lambda_{22} = \lambda_{33}$; $\beta_2 = \lambda_{\alpha\beta}$ for $\alpha \neq \beta$). To find x_D from (5.2) we used tables of this integral which were published in [12].

Figure 1 shows the dependence of Z on ϵ for T = 0 over a wide range of variation of the ratio m'/m ($W^0 = e^{-Z}$, $\epsilon = 1 - m'/m$). The ordinate is $Z\hbar\omega_{0\,max}/R'$ ($R' = \hbar^2 k^2/2m'$). The dashed curve is $Z'\hbar\omega_{0\,max}/R'$ for $\epsilon > \epsilon_c$, when the spectrum contains a discrete frequency.

From the shape of the curve it follows that, for fixed R', Z increases continually with increasing m'/m, where for $-\epsilon > 1$ the dependence rapidly
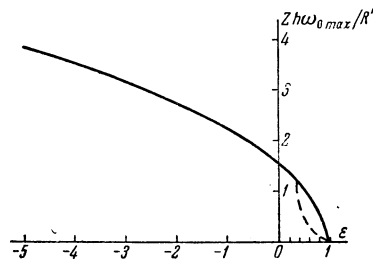
becomes a square root dependence. For $\epsilon > \epsilon_c$ the value of Z′ drops rapidly to very low values and W^0 is practically determined by Z″. All these results are in complete agreement with the general analysis given in the preceding section.

Figure 2 shows the temperature dependence of $Z\hbar\omega_{0\,max}/R'$ for different values of m'/m. The curves of Fig. 2 show very clearly the results predicted from general arguments, that there is a sharp drop in the probability for the Mössbauer effect with temperature when $m'/m \gg 1$, and that there is an equally marked drop in the temperature dependence when $m'/m \ll 1$. (When $R'/\hbar\omega_{0\,max} \ll 1$, the Bessel function I_0 in (3.15) can be replaced by unity. Then Z determines W^0 even for $m'/m < 1 - \epsilon_c$.)

The early transition to the classical limit and the vanishing of the dependence on the mass of the radiator is shown clearly in Fig. 3, where curves of
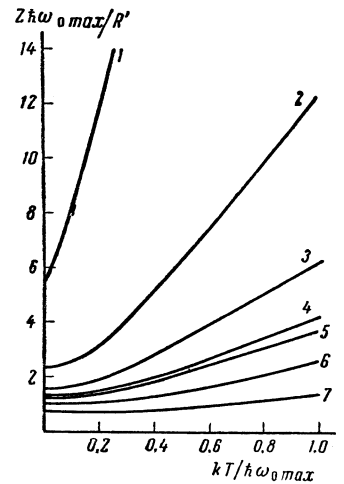
FIG. 2. Dependence of $Z\hbar\omega_{0\,max}/R'$ on temperature. Curves 1-7 correspond to the following values of m'/m: 9; 2; 1; 0.67; 0.6; 0.4; 0.2.
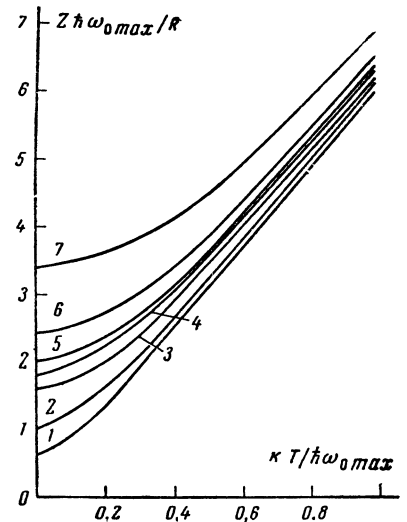
FIG. 3. Dependence of $Z\hbar\omega_{0\,max}/R$ on temperature. Curves 1-7 correspond to the following values of m'/m: 9; 2; 1; 0.67; 0.6; 0.4; 0.2.

FIG. 1

the temperature dependence are drawn, but the ordinate is $Z\hbar\omega_{0\,max}/R$, where $R = \hbar^2 k^2/2m$ is the recoil energy calculated using the mass of the free nucleus of the host.

Now let us compare these results with experiment. There are at present three experimental papers in which measurements are reported of the probability for the Mössbauer effect on an impurity nucleus in a crystal, with a sizable difference between m' and m. [3] [13-15]

1. Shirley, Kaplan, and Axel[13] measured the effect on Au^{197} nuclei introduced into relatively light lattices of Fe, Co and Ni at 4° K. With such a large mass difference (m'/m ∼ 4) the change in the force constants can be neglected and the probability for the effect should be described by the formulas given above. We have made a computation of Z = Z' for Fe and Ni, which have cubic symmetry, using (5.2). To determine the ratio of the constants for the central and noncentral interactions, $\xi = \beta_1/\beta_2$ we used tabulated values of the elastic moduli, since $\xi = c_{11}/c_{44}$. For Fe and Ni it turned out that $\xi \approx 2$.

The results of the computation are given as the ratio of Z/R' to $Z_{Fe,Ni}/R_{Fe,Ni}$ for the ideal host lattice. The values found were:

Au^{197} in Fe: $\dfrac{Z/R'}{Z_{Fe}/R_{Fe}} = 1.92$; Au^{197} in Ni: $\dfrac{Z/R'}{Z_{Ni}/R_{Ni}} = 1.86.$

The experimental results lead to values of 1.96 and 1.73, respectively for these quantities (Θ/Θ_{eff}, in the notation of [13]).

2. Heberle, Parks, and Schiffer[15] report measurements of the temperature dependence of the effect for Fe^{57} embedded in a beryllium lattice. The large mass difference (m'/m ≈ 6.3) permits the use of the limiting formulas. Unfortunately, Be has a hexagonal lattice. But when Z is small, the polycrystalline state of the sample gives rise to an averaging of the polarization vectors. Thus we can approximately use formula (4.13). Let us compute $\langle \omega_0^{-2} \rangle$. Averaging the reciprocal of the square of the frequency over a Debye spectrum, we find $\langle \omega_0^{-2} \rangle = 3/\omega_{0\,max}^2$. We note that a very similar value is obtained if one uses the model with nearest neighbor interaction and $\xi = 1$. Determining x_0 from (4.7) and computing $\bar{n}_0$, we finally get ($\hbar\omega_{0max}/k = 1000°$ K)

$$W^0\,(295°\,\text{K})/W^0\,(80°\,\text{K}) = 0.88,$$

$$W^0\,(425°\,\text{K})/W^0\,(80°\,\text{K}) = 0.81.$$

These results are in good agreement with the preliminary experimental data.

The Mössbauer effect for a light impurity nucleus (Fe^{57}) in a heavy lattice (In) was first meas-

ured by Craig, Taylor, and Nagle.[14] The authors observed an anomalously slow falloff of the probability of the effect with temperature over a wide range of temperatures for this host matrix from 1 to 300° K ($\Theta_{In} \approx 120°$ K). This result is obviously in good agreement with the general predictions of the theory (cf. I and the preceding section). Unfortunately no precise numerical results were quoted in this paper, and an indication of the approximate value of the drop in the effect over this temperature interval is given only in a footnote. We have therefore limited ourselves to relatively rough estimates of the effect for this case.

At T = 300° K, for this matrix, even when discrete frequencies are present, one definitely reaches the classical temperature limit. If as an approximation we neglect the deviation of the In lattice from cubic, then we can determine W^0 using (4.11'). Since $1 - \epsilon \approx 0.5$, we can use (4.16) to determine $\omega_D^{-2}L_D$. The moments which appear in (4.11') and (4.16) can be computed on either the nearest neighbor model or the Debye model.

The result for the ratio W^0 (300° K)/W^0(0° K) was 0.34. This value is definitely below the reduction of the probability for the effect over this temperature interval which was given in [14] by 50%. When the experimental data are obtained more accurately, we propose to make a detailed computation, based on the actual crystal structure of In. One will then be able to answer the question whether there is an effect of a change in the force constants for such a small change in the mass (m'/m ≈ 0.5).

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