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THE MÖSSBAUER EFFECT IN MONO- AND DIATOMIC CUBIC LATTICES

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We treat monatomic and diatomic cubic crystals, including both noncentral and central nearest neighbor interactions. An expression is derived for the Mössbauer effect at T = 0 and at arbitrary temperatures. We analyze the case of a diatomic lattice in which one of the atoms is the radiator. We show, in particular, that for a light radiator in a diatomic lattice the effect may be larger than for a monatomic lattice with the same Debye temperature. We find that in general for a diatomic lattice the Debye temperature does not even approximately characterize the probability of the Mössbauer effect.

INTRODUCTION

THE probability of recoilless resonant emission or absorption of γ quanta in a solid (the Mössbauer effect) depends on a quantity which is an integral characteristic of the phonon spectrum (cf. [1,2]). So far, in calculating the effect, the isotropic model and a Debye spectrum for the vibration frequencies have been used. The probability then depends on the ratio $R/k\Theta$, where R is the recoil energy and Θ the Debye temperature.

We know that the spectrum of vibrations of a real crystal is markedly different from the Debye spectrum. This immediately raises the question of the sensitivity of the Mössbauer effect to the shape of the spectrum, and whether one can use the single parameter Θ for analyzing the intensity. Of special interest in this connection are crystals with different atoms in the unit cell, where the nucleus of one of the atoms is the active nucleus.

The central point here is the analysis of the role of the optical branches, especially when there is a sizeable difference in the masses of the atoms. In particular, such an analysis should help solve the problem of whether one can obtain a sizeable effect for nuclei with a large recoil energy (i.e., relatively light nuclei) if they are incorporated into a multiatomic unit cell. We note that this problem is extremely complicated even for the case of diatomic lattices. The reason for this is principally that one must find the amplitudes of the normal modes for each atom separately, and also must include the different types of vibration branches.

In the present paper we carry out the analysis for monatomic and diatomic lattices with cubic symmetry. We use a model of the simple cubic crystal with nearest neighbor interactions (of both noncentral and central type). This model has been used in recent years for the analysis of the vibration spectra of crystals (cf. the review article [3]). An important point is that the frequency spectra obtained for the case of a monatomic lattice have all the singularities which are predicted by the general theory.[4] In the case of a diatomic lattice, the frequency distribution has an incorrect root singularity at the top of the acoustic branch and at the bottom edge of the optical branch, which is a consequence of omitting the interaction with next nearest neighbors. But a weak singularity of this type gives practically no contribution to integrals over the spectrum for the characteristic quantities, which are the only quantities of interest in the present work. Aside from this, the spectrum for the diatomic lattice has all the singularities of the general type.

1. MONATOMIC LATTICE. T = 0.

The probability for the absorption (or emission) of γ quanta in a monatomic crystal, not accompanied by any change in the phonon spectrum, is given by the expression

$$W = e^{-Z}, \tag{1.1}$$

$$Z = R\frac{v_0}{(2\pi)^3}\sum_\alpha\int d^3f\,\frac{|\mathbf{q}\mathbf{V}_\alpha(\mathbf{f})|^2}{\hbar\omega_\alpha}\,(2n_\alpha(\mathbf{f})+1), \tag{1.2}$$

where **f**, $\omega_\alpha(\mathbf{f})$ are the wave vector and frequency of a phonon corresponding to the α-th branch; $\mathbf{V}_\alpha$ is the polarization vector; n_α is the equilibrium density of phonons; **q** is a unit vector along the direction of incidence of the γ quantum, v_0 is the volume of the unit cell. For a cubic lattice at T = 0 we have

$$Z = R\frac{v_0}{(2\pi)^3}\frac{1}{3}\sum_\alpha\int d^3f\frac{1}{\hbar\omega_\alpha} = R\int\frac{1}{\hbar\omega}\psi(\omega)\,d\omega, \tag{1.3}$$

where $\psi(\omega)$ is the frequency distribution in the phonon spectrum, normalized to unity.

Let us consider a cubic crystal and include only the interaction with nearest neighbors. In this case the equations of motion for displacements along the three cubic axes are independent. We have the same secular equation for all of them:

$$m\omega^2 = 2\sum_{i=1}^{3}\gamma_i(1-\cos\varphi_i), \tag{1.4}$$

where γ_1 is the central force constant, $\gamma_2 = \gamma_3$ is the noncentral force constant; $\varphi_i = f_i a$ $(-\pi \le \varphi_i \le \pi)$.

Let us determine the frequency distribution function for such a lattice. The form of Eqs. (1.4) and the fact that all values of f_i in the space of the reciprocal lattice, and consequently all the φ_i, have the same statistical weight permits us to use the solution of the familiar random walk problem. A direct application of the general scheme gives the following expression for the distribution function (cf. the corresponding expression in [3]):

$$g(\omega^2) = \frac{1}{2\pi}\int_{-\infty}^{\infty}\exp\left\{-i\omega^2\rho + i\frac{2}{m}(\gamma_1+2\gamma_2)\rho\right\}$$

$$\times J_0\left(\frac{2\gamma_1}{m}\rho\right)J_0^2\left(\frac{2\gamma_2}{m}\rho\right)d\rho, \tag{1.5}$$

where J_0 is the Bessel function of zero order.

The function $g(\omega^2)$ is normalized by the condition

$$\int_0^\infty g(\omega^2)\,d(\omega^2) = 1. \tag{1.6}$$

From this,

$$\psi(\omega) = 2\omega g(\omega^2). \tag{1.7}$$

We substitute (1.7) and (1.5) in (1.3) and, using the fact that J_0 is an even function, go over to an integral over positive values of ρ. We interchange the order of integration and carry out the ω integration explicitly. The result is

$$Z = \frac{R}{\hbar\sqrt{\pi}}\int_0^\infty\frac{1}{\sqrt{\rho}}\sin\left(\frac{2(\gamma_1+2\gamma_2)}{m}\rho+\frac{\pi}{4}\right)J_0\left(\frac{2\gamma_1}{m}\rho\right)J_0^2\left(\frac{2\gamma_2}{m}\rho\right)d\rho \tag{1.8}$$

The expression for Z contains two independent parameters γ_1 and γ_2. In place of these we introduce two new parameters,

$$\xi = \gamma_1/\gamma_2 \tag{1.9}$$

and the Debye temperature Θ. We determine the latter by a comparison with the classical Debye formula for the low temperature specific heat of the lattice, using the dispersion law in the form (1.4). We have

$$\Theta = (\hbar/k)\,(36\pi^4\gamma_2^2\gamma_1/m^3)^{1/6}. \tag{1.10}$$

With (1.9) and (1.10), we transform (1.8) to the form

$$Z = \frac{R}{k\Theta}\left(\frac{9\pi}{2}\xi\right)^{1/6}\int_0^\infty\frac{1}{\sqrt{y}}\sin\left[(2+\xi)\,y+\frac{\pi}{4}\right]J_0(\xi y)\,J_0^2(y)\,dy. \tag{1.11}$$

Assuming R/Θ fixed, we consider Z as a function of ξ. It is easy to show that $Z \to 0$ both for $\xi \to 0$ and for $\xi \to \infty$. But at both limits Z tends to zero slowly ($\sim \xi^{1/6}$ for $\xi \to 0$ and $\sim \xi^{-1/3}$ for $\xi \to \infty$), and over a wide range of variation of ξ it changes slightly. This is shown clearly in Fig. 1,

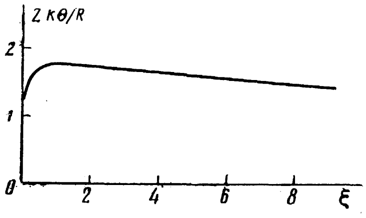

FIG. 1

where we plot the curve

$$Zk\Theta/R = f(\xi).$$

If we relate γ_1 and γ_2 to the elastic constants, it can be shown that $\xi = c_{11}/c_{44}$. From the tabulated data for c_{11} and c_{44} it follows that as a rule ξ lies in the interval 0.2 — 5. Consequently in most cases the value of Z at T = 0 will be determined mainly by the constant Θ alone.

But the actual value of $Zk\Theta/R$ is different from $^3/_2$, its value for a Debye spectrum.

2. DIATOMIC LATTICE. T = 0.

In the case of a lattice with several atoms in the unit cell, where one of them is the radiator (we give it the subscript 1), the probability for the Mössbauer effect has the form (1.1) with Z defined by the expression[2]

$$Z = R_1\frac{v_0}{(2\pi)^3}\sum_\alpha\int d^3f\,\frac{|\mathbf{q}\mathbf{V}_{1\alpha}^{*}(\mathbf{f})|^2}{\hbar\omega_\alpha(\mathbf{f})}(2n_\alpha(\mathbf{f})+1). \tag{2.1}$$

Here $V^i_{j\alpha}(f)$ is the complex amplitude which appears in the expansion of the displacement of the j-th atom in normal coordinates (cf. [2]). These amplitudes are normalized by the condition

$$\sum_{j,i} V^i_{j\alpha}(\mathrm{f})\, V^{i*}_{j\alpha}(\mathrm{f}) = 1. \tag{2.2}$$

To find the explicit form of $V^i_{j\alpha}$, we must bring in the equations of motion.

Let us consider a simple diatomic cubic lattice and include nearest neighbor interactions. The equations of motion for displacements along the three cubic axes are again independent, and the secular system of sixth order reduces to three identical pairs of equations (cf. [2]):

$$\begin{gathered} u_1^x\,[m_1\omega^2 - 2(\gamma_1 + 2\gamma_2)] + u_2^x\, 2\sum_i \gamma_i \cos\varphi_i = 0, \\ u_1^x \cdot 2\sum_i \gamma_i \cos\varphi_i + u_2^x\,[m_2\omega^2 - 2(\gamma_1 + 2\gamma_2)] = 0. \end{gathered} \tag{2.3}$$

The u_i^x are the amplitudes in the expansion in plane waves of the displacement of the i-th atom along the x axis. The rest of the notation is the same as in Sec. 1. From (2.3) we easily find the dispersion law:

$$\begin{aligned} \omega_\pm^2 &= (m_1^{-1} + m_2^{-1})(\gamma_1 + 2\gamma_2) \\ &\pm \sqrt{(m_1^{-1} - m_2^{-1})^2(\gamma_1 + 2\gamma_2)^2 + 4X^2/m_1 m_2}, \end{aligned} \tag{2.4}$$

$$X = \sum_i \gamma_i \cos\varphi_i. \tag{2.5}$$

The plus sign refers to the optical branch, the minus sign to the acoustic branch.

If we go over to normal vibrations, we can establish a relation between the amplitudes u_i^x and V_i^x:

$$u_i^x = V_i^x \sqrt{\hbar(2n + 1)/m_i\omega N}.$$

Then using one of Eqs. (2.3) and the relation (2.2), we find

$$|V_{1\alpha}^x|^2 = \frac{m_2\omega_\alpha^2 - 2(\gamma_1 + 2\gamma_2)}{2m_2\omega_\alpha^2 - 2(\gamma_1 + 2\gamma_2)(m_2/m_1 + 1)}. \tag{2.6}$$

The subscript α takes on two values, corresponding to the acoustical and optical branches. (In this model for the crystal, the polarization vector for each branch is directed exactly along one of the cubic axes).

Since (2.6) contains dependence only on ω^2, we can go over in (2.1) to an integration over frequency. We introduce the frequency distribution function $g(\omega^2)$ with the normalization (1.6). We then have for T = 0,

$$Z = \frac{4R_1}{\hbar}\int_0^\infty \frac{m_2\omega^2 - 2(\gamma_1 + 2\gamma_2)}{2m_2\omega^2 - 2(\gamma_1 + 2\gamma_2)(m_2/m_1 + 1)}\, g(\omega^2)\, d\omega. \tag{2.7}$$

The function $g(\omega^2)$ is easily related to the distribution function $G(X)$ for the quantity X of (2.5), if we use the monotonic character of the function $\omega_\pm^2(X)$ (cf. Eq. (2.4)). The same arguments as in the monatomic case lead to the expression

$$G(X) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-i\rho X} J_0(\rho\gamma_1)\, J_0^2(\rho\gamma_2)\, d\rho. \tag{2.8}$$

Changing to an integration over X in (2.7), we find

$$\begin{aligned} Z = \frac{2R_1}{\hbar}\Bigg\{ &\int_0^{\gamma_1+2\gamma_2} \frac{m_2\omega_-^2 - 2(\gamma_1 + 2\gamma_2)}{\omega_-\,[2m_2\omega_-^2 - 2(\gamma_1 + 2\gamma_2)(m_2/m_1 + 1)]}\, G(X)\, dX \\ + &\int_0^{\gamma_1+2\gamma_2} \frac{m_2\omega_+^2 - 2(\gamma_1 + 2\gamma_2)}{\omega_+\,[2m_2\omega_+^2 - 2(\gamma_1 + 2\gamma_2)(m_2/m_1 + 1)]}\, G(X)\, dX \Bigg\}. \end{aligned} \tag{2.9}$$

Here the $\omega_\pm^2$ are defined in accordance with (2.4).

Let us consider the limiting cases. Suppose first that $\epsilon = m_2/m_1 \ll 1$. We expand (2.4) in powers of ϵ. Then, keeping the first two terms of the series,

$$\omega_- = \omega_1\sqrt{1 - t^2}\,(1 - \varepsilon t^2/2), \qquad \omega_+ = \omega_1 \varepsilon^{-1/2}(1 + \varepsilon t^2/2), \tag{2.10}$$

where we have used the notation

$$\omega_1 = \frac{2}{m_1}\sum_i \gamma_i, \qquad t = \sum_i \gamma_i \cos\varphi_i \Big/ \sum_i \gamma_i. \tag{2.11}$$

The form of the expressions for $\omega_\pm$ enables us to carry out the X integration in (2.9). Omitting the computations, we give the final result for the contributions to Z from the acoustical and optical branches:

$$\begin{aligned} Z_- = \frac{2R_1}{k\Theta}\left(\frac{9\pi^4}{16}\xi(2 + \xi)^3\right)^{1/6} \Bigg\{ &\int_0^\infty J_0(y(2 + \xi))\, J_0(\xi y)\, J_0^2(y)\, dy \\ - \varepsilon &\int_0^\infty \left[J_0(y(2 + \xi)) - \frac{1}{2y(2 + \xi)} J_1(y(2 + \xi))\right] \\ &\times J_0(\xi y)\, J_0^2(y)\, dy \Bigg\}, \end{aligned} \tag{2.12}$$

$$\begin{gathered} Z_+ = \frac{2R_1}{k\Theta}\left(\frac{36}{\pi^2}\xi(2 + \xi)^3\right)^{1/6} \varepsilon^{3/2}\int_0^\infty \Bigg[\frac{2\cos(y(2 + \xi))}{y^2(2 + \xi)^2} \\ + \left(1 - \frac{2}{y^2(2 + \xi)^2}\right)\frac{\sin(y(2 + \xi))}{y(2 + \xi)}\Bigg] J_0(\xi y)\, J_0^2(y)\, dy; \\ Z = Z_- + Z_+. \end{gathered} \tag{2.13}$$

In these expressions, ξ is defined by (1.9), and the Debye temperature for the lattice is

$$\Theta = \frac{\hbar}{k}\left(\frac{288\,\pi^4\gamma_2^2\gamma_1}{(m_1+m_2)^3}\right)^{1/6}. \tag{2.14}$$

From the form of the expressions (2.12) and (2.13) it follows that the main contribution to Z comes from the acoustical branch. The contribution from the optical branches is $\sim\epsilon^{3/2}$, which is related to the fact that $\omega_+ \sim \epsilon^{-1/2}$, while the square of the amplitude $|V_1|^2$ for these branches is $\sim\epsilon$, i.e., within the framework of this model for the crystal, for $\epsilon \ll 1$ a heavy atom is hardly displaced in the optical branches.

In Fig. 2 we give the results obtained by numerical integration for the coefficients of the zeroth and first powers of ϵ in (2.12) as a function of ξ ($Zk\Theta/R_1 = A - B\epsilon$). In the interval of variation of ξ which is of practical interest, the coefficients A and B change very little.

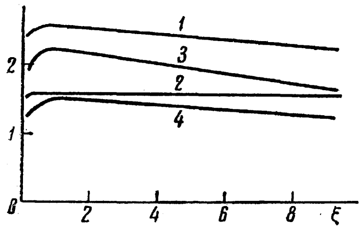

FIG. 2. Curve 1 – A(ξ), 2 – B(ξ), 3 – C(ξ), 4 – D(ξ).

Now we consider the opposite limiting case ($\epsilon \gg 1$), where the radiator is a light atom. From (2.4) we have approximately

$$\omega_- = \omega_1\varepsilon^{-1/2}(1 - t^2/2\varepsilon), \qquad \omega_+ = \omega_1(1 + t^2/2\varepsilon). \tag{2.15}$$

Substituting these values in (2.9) and carrying out the X integration, we get the expressions

$$Z_- = \frac{2R_1}{k\Theta}\left(\frac{9\pi^4}{16}\xi(2+\xi)^3\right)^{1/6}\varepsilon^{-1}\int_0^\infty\left[J_0(y(2+\xi)) - \frac{J_1(y(2+\xi))}{y(2+\xi)}\right]J_0(\xi y)\,J_0^2(y)\,dy, \tag{2.16}$$

$$Z_+ = \frac{2R_1}{k\Theta}\left(\frac{36}{\pi^2}\xi(2+\xi)^3\right)^{1/6}\left\{\varepsilon^{-1/2}\int_0^\infty\frac{\sin(y(2+\xi))}{y(2+\xi)}J_0(\xi y)\,J_0^2(y)\,dy - \varepsilon^{-3/2}\int_0^\infty\left[\frac{2\cos(y(2+\xi))}{y^2(2+\xi)^2} + \left(\frac{4}{3} - \frac{2}{y^2(2+\xi)^2}\right)\frac{\sin(y(2+\xi))}{y(2+\xi)}\right]J_0(\xi y)\,J_0^2(y)\,dy\right\}. \tag{2.17}$$

From (2.16) and (2.17) we get the important result that for $\epsilon \gg 1$ the expansion of Z in powers of $1/\epsilon$ begins with $\epsilon^{-1/2}$. As a consequence there is marked reduction in the increase of Z (decrease of the Mössbauer effect) with decreasing mass of the radiating nucleus; instead of the $1/m_1$ dependence we get a $1/\sqrt{m_1}$ dependence. Thus the Mössbauer effect for a light atom may be much more likely in a diatomic lattice than in a monatomic lattice with the same Debye temperature. Physically this gain is related to the fact that for $\epsilon \gg 1$ the light atom vibrates mainly in the optical branches. But the band of optical frequencies corresponds to a region whose characteristic frequencies are $\sqrt{\epsilon}$ times greater than the frequencies of the acoustic band.

The coefficients of the different powers of ϵ have the same order of magnitude and the same type of variation with ξ as in the preceding section. This is apparent from Fig. 2, where we give the coefficients of $\epsilon^{-1/2}$ and ϵ^{-1} in the expansions (2.17) and (2.16):

$$Zk\Theta / R_1 = C\varepsilon^{-1/2} + D\varepsilon^{-1}.$$

Figure 3 shows the dependence of $Zk\Theta/R_1$ on ϵ for a particular value of the parameter ξ ($\xi = 1$), obtained by numerical double integration of (2.9).

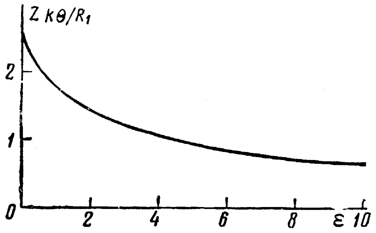

FIG. 3

This curve enables us to follow the manner in which the probability for the Mössbauer effect changes for a fixed Debye temperature, over the whole range of values for the ratio of the masses of atoms in the unit cell.

The results of this section indicate that, in the case of a diatomic lattice with an arbitrary ratio of masses, the Debye temperature does not even approximately characterize the magnitude of the Mössbauer effect. For example it is easy to find a case where a larger Debye temperature corresponds to a smaller effect.

3. MONATOMIC LATTICE. T ≠ 0.

Now let us consider the temperature dependence of the Mössbauer effect for a monatomic lattice of the same type as in Sec. 1. Transforming the general expression (1.2), we have

$$Z = R\frac{v_0}{(2\pi)^3}\int d^3f\,\frac{1}{\hbar\omega}\,\mathrm{cth}\,\frac{\hbar\omega}{2kT}. \tag{3.1*}$$

We use the transformation

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*cth = coth.

$$\frac{1}{x}\operatorname{cth} x = \int_0^\infty e^{-x^2 t}\theta(t)\,dt, \tag{3.2}$$

$$\theta(t) = \sum_{n=-\infty}^{\infty} e^{-\pi^2 n^2 t}. \tag{3.3}$$

(When we replace t by $-it'/\pi$, the function $\theta(t)$ goes over into the function $\theta_2(t')$, which coincides with the Jacobi theta function of the third kind.) Then

$$Z = \frac{R}{2kT}\frac{v_0}{(2\pi)^3}\int d^3f\int_0^\infty e^{-\beta^2\omega^2 t}\theta(t)\,dt, \tag{3.4}$$

where $\beta = \hbar/2kT$.

We substitute the expression (1.4) for ω^2 into (3.4) and integrate over the phase volume

$$Z = \frac{R}{2kT}\int_0^\infty \exp\left[-\frac{2\beta^2}{m}(\gamma_1 + 2\gamma_2)t\right]\theta(t)$$

$$\times I_0\left(\beta^2\frac{2\gamma_1}{m}t\right)I_0^2\left(\beta^2\frac{2\gamma_2}{m}t\right)dt. \tag{3.5}$$

Here I_0 is the Bessel function of pure imaginary argument.

Now we make the change of variables $\beta^2 2\gamma t/m = x$ in (3.5). Using the definitions (1.9) and (1.10) we finally get

$$Z = 2\pi\left(\frac{9\pi}{2}\xi\right)^{1/3}\frac{R}{k\Theta}\frac{T}{\Theta}$$

$$\times\int_0^\infty e^{-(2+\xi)x}\,\theta\left(4\pi\left(\frac{9\pi}{2}\xi\right)^{1/3}\frac{T^2}{\Theta^2}x\right)I_0(\xi x)\,I_0^2(x)\,dx. \tag{3.6}$$

From (3.6) it follows that $Zk\Theta/R$ is a function only of T/Θ and the parameter ξ. In Fig. 4 we show this dependence for two values of ξ. The curve for $\xi = 10$ practically coincides with the one for $\xi = 0.1$. For comparison we show the dotted curve, corresponding to a Debye spectrum. We see that the percentage variation of Z with T/Θ does not change much in the interval of values of ξ which is of most interest, and is comparatively close to the Debye case.

4. DIATOMIC LATTICE. T ≠ 0.

Using the results of Sec. 2, we find the following general expression for Z at an arbitrary temperature:

$$Z = \frac{R_1}{k\Theta}4\left(\frac{9\pi^4}{2}\xi(2+\xi)^3\frac{\varepsilon^3}{(1+\varepsilon)^3}\right)^{1/6}\sum_\alpha\int_0^1 dt\frac{F(t)\,|V_{1\alpha}(t)|^2}{\psi_\alpha(t)}$$

$$\times\operatorname{cth}\left[\frac{1}{4}\left(\frac{2}{9\pi^4}\frac{(2+\xi)^3}{\xi}\frac{(1+\varepsilon)^3}{\varepsilon^3}\right)^{1/6}\psi_\alpha(t)\frac{\Theta}{T}\right]; \tag{4.1}$$

$$F(t) = \frac{1}{\pi}\int_0^\infty \cos\rho t\,(2+\xi)\,J_0(\xi\rho)\,J_0^2(\rho)\,d\rho,$$

$$\psi_\alpha(t) = [1+\varepsilon\pm((1-\varepsilon)^2+4\varepsilon t^2)^{1/2}]^{1/2}. \tag{4.2}$$

$|V_{1\alpha}(t)|^2$ is determined by (2.6) with the value of t given by (2.11).

From the form of (4.1) it follows that $Zk\Theta/R_1$ depends on T/Θ and the parameters ξ and ϵ. The general analysis of this expression shows that the dependence on ϵ leads in the general case to a qualitative change in the way Z increases with temperature, as compared to the case of a

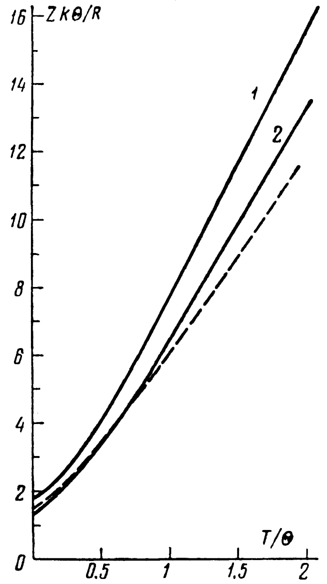

FIG. 4. Curve 1 – $\xi = 1$; curve 2 – $\xi = 0.1$.

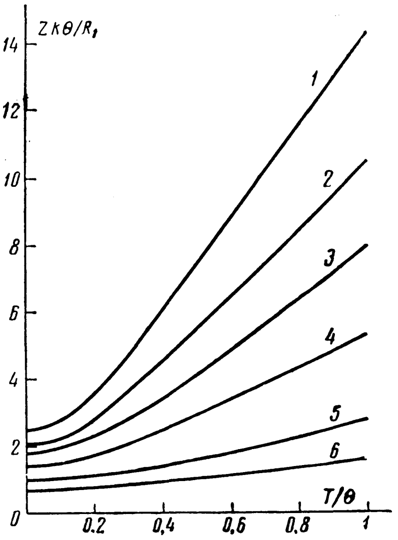

FIG. 5. Curve 1 – $\varepsilon = 0.1$; curve 2 – $\varepsilon = 0.5$; curve 3 – $\varepsilon = 1$; curve 4 – $\varepsilon = 2$; curve 5 – $\varepsilon = 5$; curve 6 – $\varepsilon = 10$.

monatomic lattice. Thus, when the radiator is the light atom and $\epsilon \gg 1$, one observes a slow variation of Z with T/Θ. As ϵ decreases the rate of increase becomes more marked, and for $\epsilon \lesssim 1$ it has a character close to that for the monatomic lattice. Such a change in the temperature dependence of the effect is related to the change in the relative importance of the optical and acoustical branches in the vibrations of the radiating atom, when there is a markedly different type of thermal excitation for the two types of phonons. (The excitation of optical phonons starts at much higher temperatures.)

Figure 5 shows curves of $Zk\Theta/R_1$ for several values of ϵ, and $\xi = 1$. We note that from our results it is clear that for an arbitrary diatomic lattice one can draw no conclusions concerning the value of the Debye temperature from the relative rate of decrease of the Mössbauer effect with temperature.

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