PHYSICAL REVIEW B VOLUME 13, NUMBER 7 1 APRIL 1976

Attraction between energy levels in the presence of relaxation

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(Received 28 July 1975)

The excited quartet states of the Mn^{++} ion in a cubic crystal are investigated from the point of view of the attraction amongst themselves as a result of the spin-phonon interaction. The diagonal relaxation probabilities and the level shift are computed. An attraction between the quartets is established theoretically and compared with experimental data of the optical absorption in Mn^{++}-doped NaCl single crystal.

INTRODUCTION

Recently Clauser[1] studied the relaxation effects in spectra using the technique of superoperators. Some general features of the first- and the second-order perturbation corrections to the eigenvalues of the superoperators were discussed in the limiting cases of fast and slow relaxations. The first-order corrections produce a broadening in the spectral lines while the second-order corrections shift the line positions. In the case of fast relaxation, the second-order correction to the frequency is[1]

$$\begin{aligned}\omega_{omn}^{(2)} &= {\sum_{\mu\nu}}' q_\nu \frac{F_{\mu\nu n} - F_{\mu\nu m}}{1+(\lambda_{\mu\nu}/\omega_{\mu\nu})^2} \\ &= {\sum_{\mu,\nu}}' q_\nu \\ &\quad \times \sum_{m',n'} \frac{|\langle \nu n|\mathcal{H}_{AB}|\mu n'\rangle|^2 - |\langle \nu m|\mathcal{H}_{AB}|\mu m'\rangle|^2}{[1+(\lambda_{\mu\nu}/\omega_{\mu\nu})^2](E_\nu - E_\mu)}, \end{aligned} \tag{1}$$

where μ, ν are the states of a system A, and m, m', n, n' are the states of the system B, and $\mathcal{H}_{AB}$ is the relaxation interaction treated as a perturbing Hamiltonian, q_ν is the probability of occupation of a state $|\nu\rangle$ separated by energy $\omega_{\mu\nu}$ from the state $|\mu\rangle$, and $\lambda_{\mu\nu} = -(W_{\mu\mu} + W_{\nu\nu})$ is the sum of relaxation rates.

In case $\lambda_{\mu\nu}/\omega_{\mu\nu} \ll 1$ and $\sum_\nu q_\nu = 1$, the fast relaxation line shifts reduce to the slow-relaxation form. However, the property of attraction between spectral lines is unchanged. For slow relaxation, the line shifts are

$$\omega_{ij}^{(2)} = \sum_{kl(\neq ij)} \frac{\langle ij|w|kl\rangle\langle kl|w|ij\rangle}{\omega_{ij}^{(0)} - \omega_{kl}^{(0)}}, \tag{2}$$

where w is the perturbation that causes relaxation and $\omega_{ij}^{(0)} = -(E_i - E_j)$ is the unperturbed energy difference between two eigenstates $|i\rangle$ and $|j\rangle$.

From the frequency-shift expression (1), it is possible to show a shift in electronic energy if we consider the relaxation of the electronic system by the transfer of energy to a phonon bath by the electron-phonon interaction.

Presently, we study the effects of spin-lattice relaxation on the spectra of the NaCl:Mn^{++} system. Clauser[1] explored the general feature that the second-order correction to eigenvalues of superoperators gives rise to attraction between the spectral lines. The general treatment is specified here for the first time, by choosing a particular system and a well-defined spin-lattice relaxation interaction. Our calculation of the second-order correction to the energies shows that attraction between energy levels in presence of relaxation does exist, i.e., the levels are indeed shifted towards each other. In this paper, "attraction" as a function of temperature is also investigated for the excited spin quartets of the Mn^{++} ion.

WAVE FUNCTIONS AND INTERACTION

The ground state of Mn^{++} ion is ^6S. There are three excited quartet states that form an interesting system to study the possibility of mutual attraction in presence of relaxation. The quartet energy levels arising due to mixing of the free ion states by the crystal field[2,3] are

$$|{}^4_i\Gamma_4\rangle = \alpha_i|{}^4P\rangle + \beta_1|{}^4F\rangle + \gamma_i|{}^4G\rangle, \tag{3}$$

where $i = 1, 2, 3$, in the increasing order of energy. The coefficients α_i, β_i, and γ_i are listed in Table I. The Hamiltonian responsible for spin-lattice relaxation[4,5] to the lowest order is

$$\begin{aligned}\mathcal{H} = \sum_j V_j Q_j = A' \sum_{ij} \{ &[Y_2^2(i) + Y_2^{-2}(i)] - \sqrt{6}\, Y_2^0(i) \\ &+ \tfrac{2}{3} i (Y_2^{-2} - Y_2^2) + \tfrac{2}{3}(Y_2^1 - Y_2^{-1}) \\ &+ \tfrac{2}{3} i (Y_2^1 + Y_2^{-1}) \} Q_j, \end{aligned} \tag{4}$$

TABLE I. Values of mixing parameters of Eq. (3).

i	α_i	β_i	γ_i
1	0.633	−0.127	0.763
2	0.761	0.285	−0.583
3	0.143	−0.950	−0.277

where V_j are the derivatives of the crystal potential terms with

$$A' = 9(\tfrac{2}{15}\pi)^{1/2} e e' \langle r^2 \rangle R^{-4} , \qquad (5)$$

in which e' is the charge of a nearest-neighboring atom, e the electronic charge, r_j the radius vector of the jth electron of the Mn^{++} ion, and R is the separation between Mn^{++} ion and the nearest neighbor. The phononic factors are described by generalized coordinates,

$$Q_j = \sum_i a_i \left(\frac{\hbar}{2M\omega_k} \right)^{1/2} (a_k + a_k^\dagger) , \qquad (6)$$

where M is the mass of the crystal, ω_k the phonon frequency of wave vector k, and $a_k^\dagger$ and a_k are the creation and destruction operators, respectively. The a_i are Van Vleck's averaging parameters given by

$$\langle a_{2i}^2 \rangle = \langle a_{3i}^2 \rangle = \circ \circ \bullet = \langle a_{6i}^2 \rangle = \tfrac{2}{15} k_l^2 R^2 ; \quad \tfrac{1}{10} k_t^2 R^2 \qquad (7)$$

for longitudinal and transverse branches, respectively, in the small-wave-vector approximation. In the Debye model the average value of the fluctuation is calculated to be

$$\langle Q_j^2 \rangle = \frac{\hbar R^2}{4\pi^2 \rho} (\tfrac{2}{3} v_l^{-5} + v_t^{-5}) \int_0^{\omega_D} [2(e^{\hbar\omega/kT} - 1)^{-1} + 1] \omega^3 d\omega . \qquad (8)$$

RELAXATION PROBABILITIES

For the calculation of the relaxation probabilities, we write the spin-lattice Hamiltonian in a slightly rearranged form:

$$\mathcal{H} = \sum_k \left(\frac{\hbar}{2M\omega_k} \right)^{1/2} |k| (a_k + a_k^\dagger) \sum_i V_i . \qquad (9)$$

In our problem, we are interested in the processes which contribute to relaxation amongst the three symmetric excited quartets separated from each other by almost 10 000 cm^{-1}. Under these circumstances, a one-phonon direct process is not possible since a phonon of such a large frequency is not available. For much the same reason, the Orbach process which is another way of looking at the direct process is not possible. However, it requires a little more than a cursory look to find that the standard Raman process is also excluded. In the later process, first a transition from a state $|b, n_{k_1}, n_{k_2}, \ldots, n_{k_{\max}}\rangle$ to a state $|c, n_{k_1} - 1, n_{k_2}, \ldots, n_{k_{\max}}\rangle$ takes place with the absorption of a phonon of wave vector k_1 and then to the final state $|a, n_{k_1} - 1, n_{k_2} + 1, \ldots, n_{k_{\max}}\rangle$ with the emission of a phonon of wave vector k_2. Apart from the matrix elements of the crystal-field factor of the interaction and the energy denominators of the intermediate state, this gives rise to phonon-matrix element of $n_{k_1}(n_{k_2} + 1)\delta(E_a - E_b - \hbar\omega_{k_1} + \hbar\omega_{k_2})$ with the double integration over k_1 and k_2 spaces. For the δ-function to give non zero probability, $E_b - E_a = \hbar\omega_{k_2} - \hbar\omega_{k_1}$. In our problem, $E_b - E_a$ is so large that the phonons of such energy differences are nonexistent. Mathematically speaking, the number density of phonons evaluated at such high energies is zero. Therefore, a two-phonon Raman process is not permissible. It is, of course, possible to consider a multiphonon process[6] with the participation of p phonons ($p > 6$ or so) with the probabilities like

$$\rho^{-p} v^{-5p} \int \int_p \cdots \int (n_{k_1} + 1)(n_{k_2} + 1) \cdots (n_{k_p} + 1) \omega_1^3 \omega_2^3 \cdots \omega_p^3 \delta(E_b - E_a - \hbar\omega_1 - \hbar\omega_2 \cdots - \hbar\omega_p) d\omega_1 d\omega_2 \cdots d\omega_p ,$$

which is always nonzero. However, such a process does not contribute to Van Vleck's relaxation time T_1 but instead, gives rise to a lifetime T_2 to the level $|b\rangle$. In general, the effective width is determined by $\lambda = 1/T_1 + 1/T_2$. In our case, it appears that the approximation $1/T_1 \ll 1/T_2$, will not upset the order of magnitude of the linewidth and therefore we restrict ourselves only to the T_2-type process, contributing to the lifetime broadening, which has not been previously considered in the excited states of Mn^{++} ion. The diagonal relaxation transition probability is given by the Fermi's

in which constants, spin matrix elements, and similar terms are left out that cannot be predicted too easily. The only process which can occur within the second-order perturbation theory is a diagonal process, beginning and ending at the same electronic state. In this process, a transition from a state $|b, n_{k_1}, n_{k_2}, \ldots\rangle$ to an intermediate state $|c, n_{k_1} - 1, n_{k_2}, \ldots\rangle$ takes place by the absorption of a phonon $\hbar\omega_{k_1}$ and then to the state $|b, n_{k_1} - 1, n_{k_2} + 1, \ldots\rangle$ by the emission of a phonon of energy $\hbar\omega_{k_2}$ so that the phonon part of the matrix element appears as $n_{k_1}(n_{k_2} + 1)\delta(\hbar\omega_{k_1} - \hbar\omega_{k_2})$, golden rule

$$W_{fi} = \frac{2\pi}{\hbar} \sum_t \left| \frac{\langle f | \mathcal{H}' | t \rangle \langle t | \mathcal{H} | i \rangle}{E_t - E_i} \right|^2 \delta (E_f - E_i) \, , \tag{10}$$

where i and f stand for initial and final states while t is an intermediate state. In the present work t has two values, consequently, there are two terms in each case, so that we write

$$W_{11} = W_{121} + W_{131} \, , \quad W_{22} = W_{212} + W_{232} \, , \quad W_{33} = W_{313} + W_{323} \, , \tag{11}$$

where the individual probabilities are defined by

$$W_{121} = \frac{2\pi}{\hbar} \left(\left| \frac{\langle {}_1\Gamma, n_k - 1, n_{k'} + 1 | \mathcal{H}' | {}_2\Gamma, n_k - 1, n_{k'} \rangle \langle {}_2\Gamma, n_k - 1, n_{k'} | \mathcal{H} | {}_1\Gamma, n_k , n_{k'} \rangle}{\Delta_{21} - \hbar\omega_k} \right|^2 + \left| \frac{\langle {}_1\Gamma, n_k - 1, n_{k'} + 1 | \mathcal{H} | {}_2\Gamma, n_k , n_{k'} + 1 \rangle \langle {}_2\Gamma, n_k , n_{k'} + 1 | \mathcal{H}' | {}_1\Gamma, n_k , n_{k'} \rangle}{\Delta_{21} + \hbar\omega_{k'}} \right|^2 \right) \delta(\hbar\omega_k - \hbar\omega_{k'}) \tag{12}$$

and like terms. In the Debye model,

$$W_{11} = \frac{9}{8\pi^3 \rho^2 v^{10}} \left(\frac{kT}{\hbar} \right)^7 \left[\int_0^{\Theta/T} \left| \left(\frac{\langle {}_1\Gamma | \Sigma V | {}_2\Gamma \rangle \langle {}_2\Gamma | \Sigma V | {}_1\Gamma \rangle}{\Delta_{21}} \right) \right|^2 2n(n+1) x^6 \, dx + \int_0^{\Theta/T} \left| \left(\frac{\langle {}_1\Gamma | \Sigma V | {}_3\Gamma \rangle \langle {}_3\Gamma | \Sigma V | {}_1\Gamma \rangle}{\Delta_{31}} \right) \right|^2 2n(n+1) x^6 \, dx \right] , \tag{13}$$

where Θ is the Debye cutoff temperature and Δ_{21} and Δ_{31} are the electronic energy separations of ${}_2\Gamma_4$ and ${}_3\Gamma_4$ with respect to ${}_1\Gamma_4$ and the approximation $\Delta_{21} \gg \hbar\omega_k$ has been used. Since the electronic energy is very much larger than the phonon energy, the phonons are extremely retarded and the lifetime of the electronic level depends on T^{-7}. In other cases, where this is not so, the reverse approximation of swift phonons ($\hbar\omega_k \gg \Delta$) leads to T^{-5} behavior. The values of matrix elements

$$\langle {}_1\Gamma | \Sigma V | {}_2\Gamma \rangle = 1.905 A' \, ,$$

$$\langle {}_1\Gamma | \Sigma V | {}_3\Gamma \rangle = 0.070 A' \, ,$$

and

$$\langle {}_2\Gamma | \Sigma V | {}_3\Gamma \rangle = 1.03 A' \, ,$$

are calculated using the Racah algebra[2,7] as described in Ref. 2. Substituting various parameters for NaCl:Mn^{++}, (13) becomes

$$W_{11} = 0.12 T^7 \int_0^{\Theta/T} (e^x - 1)^{-2} e^x x^6 \, dx , \tag{14}$$

similarly,

$$W_{22} = 0.14 T^7 \int_0^{\Theta/T} (e^x - 1)^{-2} e^x x^6 \, dx , \tag{15}$$

$$W_{33} = 0.02 T^7 \int_0^{\Theta/T} (e^x - 1)^{-2} e^x x^6 \, dx . \tag{16}$$

At 1 °K, the above probabilities reduce to

$$W_{11} = 86.7 \text{ sec}^{-1} , \quad W_{22} = 100.3 \text{ sec}^{-1} , \quad W_{33} = 13.6 \text{ sec}^{-1} , \tag{17}$$

and at 4.2 °K,

$$W_{11} = 1.99 \times 10^6 \text{ sec}^{-1} , \tag{18}$$

$$W_{22} = 2.31 \times 10^6 \text{ sec}^{-1} , \tag{19}$$

$$W_{33} = 3.14 \times 10^5 \text{ sec}^{-1} , \tag{20}$$

and at room temperature they become of the order of 10^{13}–10^{14} sec^{-1}. Since our energy-level separations are

$$\omega_{12} = E_{{}_2\Gamma} - E_{{}_1\Gamma} = 3 \times 10^{15} \text{ sec}^{-1} , \quad \omega_{13} = E_{{}_3\Gamma} - E_{{}_1\Gamma} = 5 \times 10^{15} \text{ sec}^{-1} , \quad \omega_{23} = E_{{}_3\Gamma} - E_{{}_2\Gamma} = 1.8 \times 10^{15} \text{ sec}^{-1} , \tag{21}$$

so that

$$\frac{\lambda_{ij}}{\omega_{ij}} = \frac{-(W_{ij} + W_{ji})}{\omega_{ij}} < 1 , \tag{22}$$

indicating that the widths are smaller than the separations. Consideration of (22) in (1) shows that for all the levels the ratio of relaxation rate to the frequency difference is such that the ratio $\lambda_{\mu\nu} / \omega_{\mu\nu}$ being small compared with unity, can be ignored in the denominator of (1). Thus the relaxation amongst various levels can be treated as a case of slow relaxation.

ATTRACTION BETWEEN QUARTET LEVELS

In this section simplified expressions for the attraction, i.e., the shifts of levels defined by (3) are given. We use the spin-lattice Hamiltonian (4) and calculate the correction to the energy in second order. The changes in the energy of the levels are

$$\Delta E_{{}_1\Gamma_4} = \langle {}_1\Gamma_4 | \mathcal{H} | {}_2\Gamma_4 \rangle \langle {}_2\Gamma_4 | \mathcal{H}' | {}_1\Gamma_4 \rangle / (E_{{}_2\Gamma_4} - E_{{}_1\Gamma_4}) + \langle {}_1\Gamma_4 | \mathcal{H} | {}_3\Gamma_4 \rangle \langle {}_3\Gamma_4 | \mathcal{H}' | {}_1\Gamma_4 \rangle / (E_{{}_3\Gamma_4} - E_{{}_1\Gamma_4}), \tag{23}$$

$$\Delta E_{{}_2\Gamma_4} = \langle {}_2\Gamma_4 | \mathcal{H} | {}_1\Gamma_4 \rangle \langle {}_1\Gamma_4 | \mathcal{H}' | {}_2\Gamma_4 \rangle / (E_{{}_1\Gamma_4} - E_{{}_2\Gamma_4}) + \langle {}_2\Gamma_4 | \mathcal{H} | {}_3\Gamma_4 \rangle \langle {}_3\Gamma_4 | \mathcal{H}' | {}_2\Gamma_4 \rangle / (E_{{}_3\Gamma_4} - E_{{}_2\Gamma_4}), \tag{24}$$

$$\Delta E_{{}_3\Gamma_4} = \langle {}_3\Gamma_4 | \mathcal{H} | {}_2\Gamma_4 \rangle \langle {}_2\Gamma_4 | \mathcal{H}' | {}_3\Gamma_4 \rangle / (E_{{}_2\Gamma_4} - E_{{}_3\Gamma_4}) + \langle {}_3\Gamma_4 | \mathcal{H} | {}_1\Gamma_4 \rangle \langle {}_1\Gamma_4 | \mathcal{H}' | {}_3\Gamma_4 \rangle / (E_{{}_1\Gamma_4} - E_{{}_3\Gamma_4}). \tag{25}$$

Here the phonon contribution to shifts is contained in the Hamiltonian (4). Again, the matrix elements are found using the seniority scheme of Racah as described by Blume and Orbach, and the constants of Table I are employed. The energy shifts for each level in (23)–(25) are found to be the sum of two contributions. For the levels ${}_1\Gamma_4$ and ${}_3\Gamma_4$, the dominant attraction arises from ${}_2\Gamma_4$ whereas the second contribution is smaller by a factor of 10^{-3}. However for the level ${}_2\Gamma_4$, the attraction from ${}_1\Gamma_4$ and ${}_3\Gamma_4$ is of the same order of magnitude and therefore both contributing levels are important. The energy shift of ${}_2\Gamma_4$ is the resultant of two oppositely directed contributions from the levels ${}_1\Gamma_4$ and ${}_3\Gamma_4$ and the attraction due to ${}_1\Gamma_4$ is somewhat stronger than that of ${}_3\Gamma_4$ as indicated by the negative sign of the energy shift. It may be noted that for the lowest level, both the energy denominators are positive whereas for the top level, both the denominators are negative and for the intermediate level, one of the denominators is positive and one negative.

For[5] $\rho = 2.168$ g/cm^3, $R = 2.78 \times 10^{-8}$ cm, $v_l = 4.78 \times 10^5$ cm/sec, $v_t = 2.43 \times 10^5$ cm/sec, $\langle r^2 \rangle = 1.584$ a.u., $e = e'$, the expressions for the attraction after ignoring negligible terms are

$$\Delta E_{{}_1\Gamma_4} = 4 \times 10^{-8}\Theta^4 + 3 \times 10^{-7} T^4 \int_0^{\Theta/T} (e^x - 1)^{-1} x^3\, dx, \tag{26}$$

$$\Delta E_{{}_2\Gamma_4} = -1.5 \times 10^{-8}\Theta^4 - 1.2 \times 10^{-7} T^4 \int_0^{\Theta/T} (e^x - 1)^{-1} x^3\, dx, \tag{27}$$

$$\Delta E_{{}_3\Gamma_4} = -2.3 \times 10^{-8}\Theta^4 - 1.8 \times 10^{-7} T^4 \int_0^{\Theta/T} (e^x - 1)^{-1} x^3\, dx. \tag{28}$$

These energy shifts have temperature-dependent as well as temperature-independent parts. The temperature-independent contribution is due to the zero-point vibration, whereas the temperature-dependent part in the Debye model varies as T^4 at low temperatures owing to the approximations employed (Appendix A).

The excited quartet states of the Mn^{++} ion in NaCl have been measured in the optical absorption[8,9] at the liquid-nitrogen temperature and the room temperature. So far in our theory there is no adjustable parameter. We now allow the Debye cutoff to be the only adjustable parameter. For a value of $\Theta = 150$ K, for which the best agreement occurs, we show our calculated quartets along with the experimental values in Fig. 1. Indeed, Clauser's attraction between the quartets is very clearly displayed, and as far as we know is demonstrated here for the first time in electronic spectra.

CONCLUSIONS

In the present work we have discovered that the spin-lattice-relaxation interaction causes an attraction between the energy levels. The attraction is inversely proportional to the separation between the levels themselves. Clauser's possibility of attraction of spectral lines using superoperators is explained using perturbation treatment and experimental evidence is provided for the first time. In addition the diagonal self-energy process for the excited quartets is calculated here for the first time.

APPENDIX A

In the expressions (26)–(28) we have employed a long-wavelength approximation (7) for the lattice phonons. There has been a considerable amount of discussion[10] on the validity of this approximation, and though "more accurate" calculations have been performed, it is not answered as to how much is the difference. Therefore, in this appendix, we

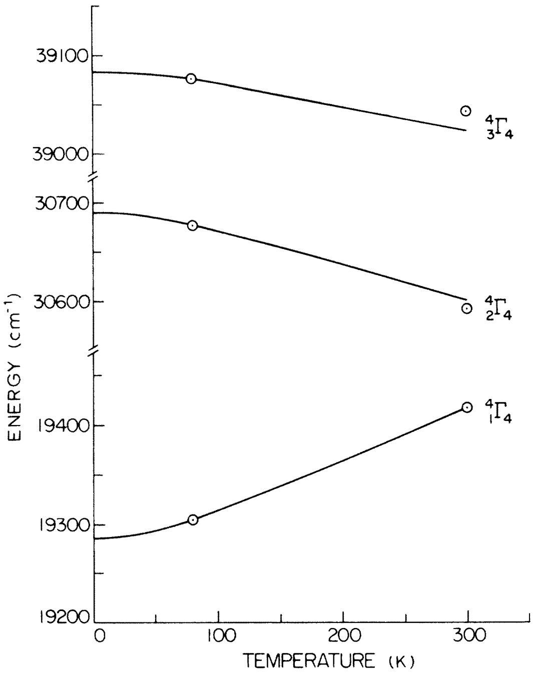

FIG. 1. Excited quartets of the Mn^{++} ion in a cubic crystal are shown to come closer as a function of temperature. The curves are calculated with a single adjustable parameter $\Theta = 150$ K. The circles are the experimental observations from the optical absorption.

indicate the calculation without the approximation and compute the nature of the approximation itself. Avoiding the approximation, the average values of the squares of Van Vleck's normal coordinates are

$$\langle Q_2^2\rangle_l = \langle Q_3^2\rangle_l = \frac{1}{3}\sum_i (2n_i + 1)[1 - j_0(2k_iR) - 3j_2(\sqrt{2}\,k_iR)], \tag{A1}$$

$$\langle Q_4^2\rangle_l = \langle Q_5^2\rangle_l = \langle Q_6^2\rangle_l = \frac{1}{3}\sum_i (2n_i + 1)[1 - j_0(2k_iR) - j_2(2k_iR) + 3j_2(\sqrt{2}\,k_iR)]$$

for the longitudinal component of the phonon wave vector k, and

$$\langle Q_2^2\rangle_t = \langle Q_3^2\rangle_t = \frac{1}{3}\sum_i (2n_i + 1)[1 - j_0(2k_iR) - j_2(2k_iR) + \tfrac{1}{2}j_2(\sqrt{2}\,k_iR)], \tag{A2}$$

$$\langle Q_4^2\rangle_t = \langle Q_5^2\rangle_t = \langle Q_6^2\rangle_t = \frac{1}{3}\sum_i (2n_i + 1)[1 - j_0(2k_iR) + \tfrac{1}{2}j_2(2k_iR) - \tfrac{3}{2}j_2(\sqrt{2}\,k_iR)]$$

for the transverse component. For changing the sum into integration one has to add two transverse and one longitudinal component taking into account that the longitudinal velocity is slightly faster than the transverse velocity and the density of states as determined by the Debye model. If the transverse

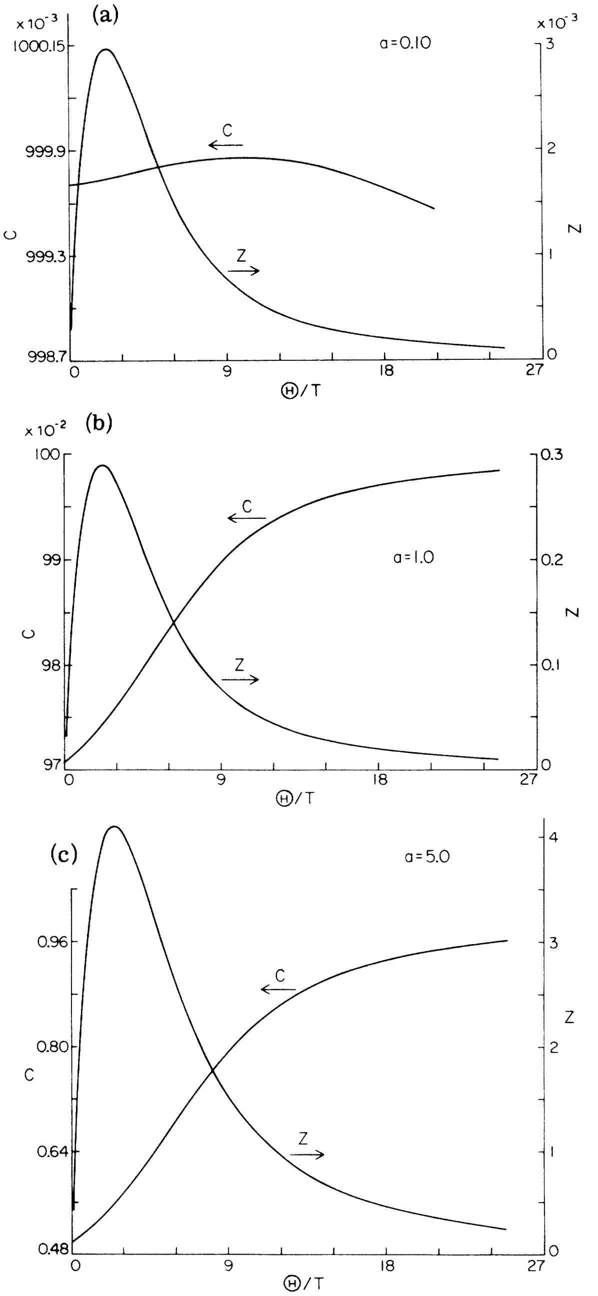

FIG. 2. Correction factor C and the function Z avoiding the long-wavelength approximation as a function of Θ/T and for (a) $a = 0.1$, (b) $a = 1.0$, and (c) $a = 5.0$. This calculation shows that the long-wavelength approximation is not serious in view of adjustability of a parameter.

and the longitudinal phonon wave vectors are assumed equal, all of the second-order Bessel functions vanish and instead of the integral

$$L = b^2 \int_0^{\Theta/T} \frac{x^3\,dx}{e^x - 1}, \tag{A3}$$

we get

$$Z = 6 \int_0^{\Theta/T} (e^x - 1)^{-1} [1 - j_0(bx)]\, x\,dx, \tag{A4}$$

where

$$b = a(T/\Theta), \quad a = (k/\hbar)(\Theta/v)R. \tag{A5}$$

The solution of (A3) has been discussed by Herberle.[11] However (A4) cannot be solved too easily and must be machine integrated. As the value of a depends on the lattice constant, the Debye cutoff, and the sound velocity, it will acquire somewhat different values in different materials. We define a correction factor

$$C = Z/L, \tag{A6}$$

with which the approximate value (A3) must be multiplied to yield the "improved" result. In Figs. 2(a)–2(c), we show numerically integrated values.

Indeed, the correction (A6) is not serious unless the value of a given by (A5) is unfavorably large, which does not happen in our problem. We have also numerically integrated the Bessel functions of the second order involved in (A2) and find that their effect is small and indeed vanishes for isotropic phonons.

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[*] Supported by the Council of Scientific and Industrial Research, New Delhi.

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