

expected for an elastically isotropic medium and with only longitudinal acoustic phonons participating in the phonon-drag effect, but since neither condition applies to germanium, the result is fortuitous. A curvature in the plot of $\log Q_\theta$ versus $\log T$ is not out of the question and could be in a direction to either increase or decrease b and d .

Because of the possibility that b and d have been overestimated as a result of mass changes or non-constant exponent in the phonon drag, this experiment is not inconsistent with b and d equal to zero, although we believe this is unlikely. In any case, $(b+d)$ is much smaller than expected. Since the energy levels in metals should also be temperature dependent as a result of temperature-induced disorder in the crystal lattice,²⁵

²⁵ S. F. Edwards, Proc. Roy. Soc. (London) **A267**, 518 (1962).

the point of view developed in this paper would predict new contributions to the thermoelectric power in metals also. In fact, as pointed out in Sec. II, any interaction that scatters electrons will also change their energies to some extent and may contribute to the thermoelectric power through this energy change as well as through the usual effect on $j(E)$.

ACKNOWLEDGMENTS

We wish to thank the David Sarnoff Laboratory of the Radio Corporation of America for supplying us with samples, and especially Drs. S. Christian and B. Abeles and Mr. J. Gannon for their interest. We also wish to thank Professors B. Serin and P. Weiss for their helpful comments.

Phonon Spectrum and Specific Heats of Copper Using Baily's Effective Matrix Element

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(Received 18 May 1965)

Phonon spectrum of copper has been calculated by using Baily's total effective matrix element for the electron-phonon interaction. The results show that the agreement with the neutron-scattering results, specific heat, and elastic constant is better than that obtained with Toya's matrix element.

1. INTRODUCTION

AMONG the face-centered-cubic metals, copper has been a favorite subject of study of both the theoretical and experimental workers. De Launay^{1,2} used his elastic-force model to explain the temperature dependence of the specific heats. The detailed calculations based on this model were carried out by Srivastava.³ Though the specific-heat results are in fair agreement with the experiment, the vibration spectra show noticeable disagreement with the experimental dispersion curves obtained from neutron diffraction (Cribier *et al.*,⁴ Sinha and Squires⁵). Sharma and Joshi⁶ have made use

of their own model for computing the specific heats and the vibration spectra in the three principal directions. The agreement with the experimental dispersion curves is not very satisfactory and their calculated θ - T curve shows a disagreement of about 5% with the experimental curve. Shukla^{7,8} has tried Krebs's model.⁹ The agreement with the dispersion curves as well as the specific heats is found to be excellent.

The elastic-force models, in spite of their success, are intellectually unsatisfactory because they ignore the interactions actually present in metals. Dayal and Tripathi¹⁰ and Toya¹¹ have developed methods based on actual metallic interactions. The vibration frequencies are calculated from a secular determinant whose elements are the sum of three coupling coefficients: $[xy]^C$, $[xy]^R$, and $[xy]^E$. The first two refer to the contributions of the Coulomb and non-Coulomb

¹ J. De Launay, J. Chem. Phys. **21**, 1975 (1953).

² J. De Launay, *Solid State Physics* (Academic Press Inc., New York, 1956), Vol. 2, p. 220.

³ P. L. Srivastava, Phys. Status Solidi **2**, 713 (1962).

⁴ D. Cribier, B. Jacrot, and D. Saint-James, *Proceedings of the Symposium on Inelastic Scattering of Neutrons in Solids and Liquids, Vienna, 1960* (International Atomic Energy Agency, Vienna, 1961), p. 549.

⁵ S. K. Sinha and G. L. Squires, *Proceedings of the Conference on Lattice Dynamics, Copenhagen, 1963* (Pergamon Press, Inc., New York, 1963), p. 53.

⁶ P. K. Sharma and S. K. Joshi, J. Chem. Phys. **39**, 2633 (1963).

⁷ M. M. Shukla, Phys. Status Solidi **7**, K11 (1964).

⁸ M. M. Shukla, Phys. Status Solidi **8**, 475 (1965).

⁹ K. Krebs, Phys. Letters **10**, 12 (1964).

¹⁰ B. Dayal and B. B. Tripathi, J. Chem. Phys. **36**, 3002 (1962).

¹¹ T. Toya, J. Res. Inst. Catalysis, Hokkaido Univ. **6**, 161 (1958).

interactions, respectively, while the third owes its origin to the conduction electrons. Toya has calculated all three terms rigorously. Dayal and Tripathi, however, calculated only the first two in a rigorous manner. The electronic coupling coefficient is derived phenomenologically from the electronic bulk modulus. While the phonon dispersion curves in the case of copper show a fair but not exact agreement with the experimental data, θ shows a deviation of about 5%.

Toya has considered the contribution of the conduction electrons towards the elements of the dynamical matrix in a fundamental way. His work is essentially an extension of Bardeen's work,¹² in the respect that he has included the effect of exchange on the screening of the ionic perturbation. He has used Slater's¹³ simplification for the exchange potential. Expanding the potential of the ions in a power series of the normal coordinates he has determined the adiabatic potential energy of the valence electrons and hence the electronic coupling coefficient. It is well known that the Hartree-Fock equation, on which Toya's work is based, does not take into account the correlation of the antiparallel spins. It also does not give the correct density of states. Toya corrected for these effects by introducing correction terms dependent on the two parameters B and D/D_0 . B takes into account the effect of correlation of both spins on the screening of the ionic perturbation. Its numerical value was determined in such a way that the expression for the compressibility became identical with the one given earlier by Seitz.¹⁴ D and D_0 are the densities of states with and without the exchange correlation, respectively.

Woods *et al.*¹⁵ pointed out that except for the T_1 branch of the [110] direction their experimental dispersion curves for sodium obtained from neutron scattering showed good agreement with the results of Toya's¹⁶ calculations. Srivastava and Srivastava¹⁷ made a preliminary computation of the specific heats of sodium and potassium on the basis of Toya's theory and found that the theoretical C_v are considerably higher than the experimental values. The present authors¹⁸ made a fresh computation of the vibration spectrum and the specific heats of sodium on this theory by making a small change in one of the functions occurring in the expression for the coupling coefficient. In this way they were able to obtain good agreement with the experimental results under both heads. R. S.

Srivastava and Dayal¹⁹ also obtained good agreement in the case of the specific heat of potassium by making a similar modification. Toya^{20,21} himself calculated the dispersion curves of copper in the three symmetry directions. A comparison of these with the later neutron results (Cribier *et al.*⁴ and Sinha and Squires⁵) shows an unsatisfactory agreement.

Bailyn²² has also calculated the effective matrix element by the Hartree-Fock method. The electronic coupling coefficients on this theory can be calculated by replacing Toya's matrix element with that of Bailyn in the energy expression derived by Toya. Bailyn's matrix element does not involve the uncertain parameters B and D/D_0 and was used by the present authors²³ in an earlier paper to calculate the specific heats and the vibration spectrum of sodium. The frequencies as well as the specific heats were found to be nearly the same as in Toya's theory in its modified form (Dayal and Srivastava¹⁸). In view of this agreement it was thought that the discrepancy found in Toya's theory for copper might be removed by using Bailyn's expression for the matrix element. In the present paper we have made use of the ionic perturbation determined by Bailyn²² for the vibration frequencies of copper. Our results indicate that the use of Bailyn's effective matrix element leads to a remarkable improvement in the agreement with the neutron-diffraction results for the vibration spectra. The agreement is quite satisfactory in the case of specific heats and elastic constants also.

2. ELEMENTS OF THE SECULAR DETERMINANT

Proceeding in the usual way the secular determinant for the determination of the vibration frequencies may be written as

$$|M - m\omega^2 I| = 0, \tag{1}$$

where M is the dynamical matrix, m the mass of the ion, ω the circular frequency and I the unit matrix of order three. An element $[xy]$ of the dynamical matrix M can be written as the sum of three coupling coefficients $[xy]^C$, $[xy]^R$, $[xy]^B$ which are calculated as below:

$$[xy]^C$$

The expressions for these have been derived by Kellermann.²⁴ For a mesh of 1000 points in the Brillouin zone they can be read directly from his paper.

¹² J. Bardeen, Phys. Rev. **52**, 688 (1937).
¹³ J. C. Slater, Phys. Rev. **81**, 385 (1951).
¹⁴ F. Seitz, *Modern Theory of Solids* (McGraw-Hill Book Company, Inc., New York, 1940).
¹⁵ A. D. B. Woods, B. N. Brockhouse, R. H. March, A. T. Stewart, and R. Bowers, Phys. Rev. **128**, 1112 (1962).
¹⁶ T. Toya, J. Res. Inst. Catalysis, Hokkaido Univ. **6**, 183 (1958).
¹⁷ P. L. Srivastava and R. S. Srivastava, Proc. Phys. Soc. (London) **81**, 586 (1963).
¹⁸ B. Dayal and P. L. Srivastava, Proc. Roy. Soc. (London) **A277**, 183 (1964).

¹⁹ R. S. Srivastava and B. Dayal, Progr. Theoret. Phys. (Kyoto) **31**, 167 (1964).
²⁰ T. Toya, Progr. Theoret. Phys. (Kyoto) **20**, 974 (1958).
²¹ T. Toya, J. Res. Inst. Catalysis (Kyoto), Hokkaido Univ. **9**, 178 (1961).
²² M. Bailyn, Phys. Rev. **117**, 974 (1960).
²³ B. Dayal and P. L. Srivastava, Proc. Roy. Soc. (London) **A283**, 394 (1965).
²⁴ E. W. Kellermann, Phil. Trans. Roy. Soc. (London) **A238**, 513 (1940).

$$[xy]^R$$

These are given by

$$[xy]_{x=y}^R = 2(\phi'' + \phi') [2 - C_1(C_2 + C_3) + 4\phi'(1 - C_2C_3)], \quad (2)$$

$$[xy]_{x \neq y}^R = 2(\phi'' - \phi') S_1 S_2,$$

where $C_i = \cos(r_0 q_i)$ and $S_i = \sin(r_0 q_i)$; q_i are the components of the wave vector \mathbf{q} with $q = 2\pi/\lambda$. ϕ'' and ϕ' are defined by

$$\phi'' = d^2\phi/dR^2 \quad \text{and} \quad \phi' = (1/R)d\phi/dR \quad (3)$$

evaluated at the equilibrium position of the ions. ϕ is the non-Coulomb potential assumed to be given by the Born-Mayer²⁵ expression,

$$\phi = A e^{-R/\rho} \quad (4)$$

R is the interionic distance. A and ρ are constants.

$$[xy]^E$$

The expressions for the electronic coupling coefficients may be easily found out by determining the adiabatic potential energy of the valence electrons in terms of the normal coordinates. Its expression has been derived by Toya and involves the matrix elements V_{ih_+} and V_{ih_-} for the perturbation δV_i in the ionic potential V_i and V_{ph_+} , V_{ph_-} for the perturbation δV_p in the total potential V_p . These are defined as follows:

$$V_{ih_+} = \int \psi(\mathbf{K} + \mathbf{q} + \mathbf{K}_h, \mathbf{r})^* \delta V_i \psi(\mathbf{K}, \mathbf{r}) d\mathbf{r} \quad (5)$$

and

$$V_{ph_+} = \int \psi(\mathbf{K} + \mathbf{q} + \mathbf{K}_h, \mathbf{r})^* \delta V_p \psi(\mathbf{K}, \mathbf{r}) d\mathbf{r}. \quad (6)$$

V_{ih_-} and V_{ph_-} are also defined in similar manner by replacing plus sign in the function by a negative sign. Here ψ is the wave function of an electron in state \mathbf{K} and position \mathbf{r} . \mathbf{q} is the phonon wave vector which perturbs the lattice and \mathbf{K}_h is the reciprocal lattice wave vector. Assuming V_{ih_+} to be known, V_{ph_+} may be found in terms of V_{ih_+} . Bardeen¹² determined a relation between the two in the Hartree approximation as

$$V_{ph_+} = S V_{ih_+}, \quad (7)$$

where

$$S = \left\{ 1 - \frac{8\pi e^2}{|\mathbf{q} + \mathbf{K}_h|^2} \sum_K [E(\mathbf{K}) - E(\mathbf{K} + \mathbf{q} + \mathbf{K}_h)]^{-1} \right\}^{-1}; \quad (8)$$

$E(\mathbf{K})$ is the energy of an electron in the state \mathbf{K} . In the above expression the effect of exchange has not been considered. Toya¹¹ included the exchange effect in the framework of Bardeen's theory of screening of the ionic perturbation. He used Slater's simplification for the

²⁵ M. Born and J. Mayer, Z. Physik, 75, 1 (1932).

exchange potential and found S as

$$S = \left\{ 1 - \left[\frac{8\pi e^2}{|\mathbf{q} + \mathbf{K}_h|^2} - \frac{5}{6} \left(\frac{2e^2}{N} \right) \left(\frac{3N}{8\pi} \right)^{\frac{1}{3}} \right] \times \sum_K [E(\mathbf{K}) - E(\mathbf{K} + \mathbf{q} + \mathbf{K}_h)]^{-1} \right\}^{-1}, \quad (9)$$

where N is the number of ions per unit volume. It differs from Bardeen's S in respect of the second term in the first bracket in the denominator of (9).

The neglect of the correlation of the other part of the spin leads to serious troubles as regards density of states near the Fermi surface. To include the effect of this part of the spin, Toya resorted to two empirical modifications. The above expression for S was modified by replacing the second term in the first bracket by $2B\pi e^2/K_F^2$ where B is a parameter assumed independent of \mathbf{q} and K_F is the Fermi radius. Further the second bracket in Eq. (9) was multiplied by D/D_0 to take into account the reduction of density of states near the Fermi surface. The expression used for $E(\mathbf{K})$ was the same as obtained in the Hartree approximation.

Bailyn²² also extended Bardeen's work by including the effect of exchange on the screening of the ionic perturbation. Following Bohm and Pines,²⁶ the Coulomb interaction $e^2/|\mathbf{r}_1 - \mathbf{r}_2|$ was expanded into two parts as follows:

$$\frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} = 4\pi e^2 \left(\sum_{K'' < K_C} \frac{\exp i\mathbf{K}'' \cdot (\mathbf{r}_1 - \mathbf{r}_2)}{K''^2} + \sum_{K'' > K_C} \frac{\exp i\mathbf{K}'' \cdot (\mathbf{r}_1 - \mathbf{r}_2)}{K''^2} \right), \quad (10)$$

where $K_C = \beta K_F = 0.353 (r_s/a_0)^{1/2} K_F$, r_s is the radius of the atomic polyhedron, a_0 is the Bohr radius and \mathbf{r}_1 and \mathbf{r}_2 are the position vectors of two interacting electrons. The collective approach of Bohm and Pines was used so that the electrons below a certain value behave like a plasma while the rest can be considered as independent electrons. Under the circumstances the exchange energy comes from the second term. According to Bailyn this corresponds to a simple modification of the exchange hole around the electron. He obtained an integral equation involving V_{ph_+} and V_{ih_+} . The solution of this equation in the zeroth approximation gives

$$V_{ph_+} = S V_{ih_+}, \quad (11)$$

where

$$S = \left\{ 1 + [(9\pi/4)^{1/3} \pi]^{-1} (r_s/a_0) [F(u) - G(u, h)]/u^2 \right\}^{-1}, \quad (11a)$$

where

$$F(u) = \frac{f(u)}{p} + \frac{q}{p} \left[\frac{1-u^4}{4u} \ln \left| \frac{1+u}{1-u} \right| + \frac{u^2}{2} - \frac{3}{2} \right], \quad (12)$$

$$f(u) = 0.5 + [(1-u^2)/(4u)] \ln |(1+u)/(1-u)|, \quad (13)$$

²⁶ D. Bohm and D. Pines, Phys. Rev. 92, 609 (1953).

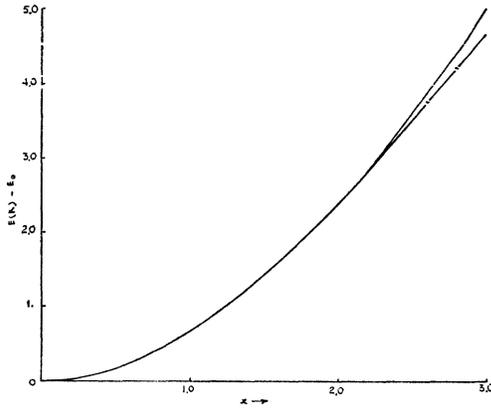


FIG. 1. $E(K) - E_0$ versus x . —, true energy curve obtained from Eq. (15); ----, true energy curve approximated by Eq. (14).

and $G(u, h)$ is a complicated function of u given by Eq. A(22) of Bailyn.²² $u = |\mathbf{q} + \mathbf{K}_h|/2K_F$. p and q are parameters obtained by approximating the energy function assumed to be of the form

$$E(\mathbf{K}) = E_0 + (\hbar^2 K_F^2 / 2m)(px^2 - pqx^4) \quad (14)$$

with the true energy function

$$E(\mathbf{K}) = \hbar^2 K^2 / 2m + W(\mathbf{K}), \quad (15)$$

where

$$W(\mathbf{K}) = -\frac{4\pi e^2}{8\pi^3} \int_{|\mathbf{K}-\mathbf{K}'| > K_C} \frac{d\mathbf{K}'}{|\mathbf{K}-\mathbf{K}'|^2} \quad (16)$$

$$= -\frac{2e^2}{\pi} K_F h(x),$$

where

$$x = K/K_F$$

and

$$h(x) = f(x) - \beta, \quad 0 < x < 1 - \beta$$

$$= \frac{1}{4} - \frac{\beta}{2} + \frac{1-x^2}{4x} \ln \frac{1+x}{\beta} + \frac{3x^2-1+\beta^2}{8x}, \quad 1-\beta < x < 1+\beta$$

$$= f(x), \quad 1+\beta < x < 3. \quad (17)$$

Taking the expression for adiabatic potential energy from Toya's paper and using V_{ph_+} given by (11) and the energy expression for $E(\mathbf{K})$ from (14), the electronic coupling coefficient assumes a form given as follows:

$$[xy]^E = N e^2 \left[\frac{4\pi}{3} \delta_{xy} - 4\pi \sum_h \frac{(q_x + K_{hx})(q_y + K_{hy})}{|\mathbf{q} + \mathbf{K}_h|^2} \times \frac{8\pi N e^2}{|\mathbf{q} + \mathbf{K}_h|^2} \frac{3F(u)^2}{4W_F} \right], \quad (18)$$

$$\delta_{xy} = 1 \quad \text{for } x = y$$

$$= 0 \quad \text{for } x \neq y,$$

S is given by (11a) and

$$G(u) = [1 + (V(r_s) - E_0)((u^2 K_F^2) / (\pi N e^2))] g(x). \quad (19)$$

$V(r_s)$ is the potential at the surface of the Seitz polyhedron and E_0 is the energy of the electron in the lowest state.

$$g(x) = 3(\sin x - x \cos x) / x^3; \quad x = |\mathbf{q} + \mathbf{K}_h| r_s, \quad (20)$$

$$W_F = \hbar^2 K_F^2 / 2m. \quad (21)$$

3. FREQUENCY SPECTRA AND SPECIFIC HEATS OF COPPER

For the purpose of the calculation of the vibration frequencies and specific heats, the reciprocal space has been divided into miniature cells whose axes are 1/10 of the axes of the ordinary reciprocal cell. This gives 1000 points of the miniature cell lying within the first Brillouin zone. From symmetry considerations the secular determinant (1) has to be solved for 48 representative points lying within the 1/48 part of the zone irreducible under the symmetry operations which leave the roots of the secular determinant unchanged. The electrostatic coupling coefficients for each of them were taken from the work of Kellermann²⁴ and Dayal and Tripathi.²⁷ The repulsive coupling coefficients were calculated from Eq. (2). Toya has used the following values for the parameters in the Born-Mayer expression (4):

$$A = 0.156 e^{2r_0/\rho} \text{ eV}; \quad \rho = r_0 / 7.148,$$

where r_0 is half the lattice constant. In addition to these values he selected the following values for the various parameters in the electronic coupling coefficient:

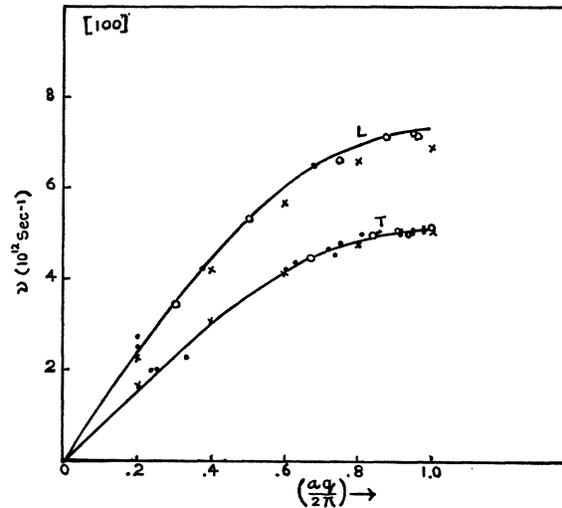


FIG. 2. Dispersion curve for copper for [100] direction. —, authors theoretical; X, Toya's theoretical; O, experimental (Cribier *et al.*); ●, experimental (Sinha and Squires).

²⁷ B. Dayal and B. B. Tripathi, Proc. Phys. Soc. (London) 77, 303 (1961).

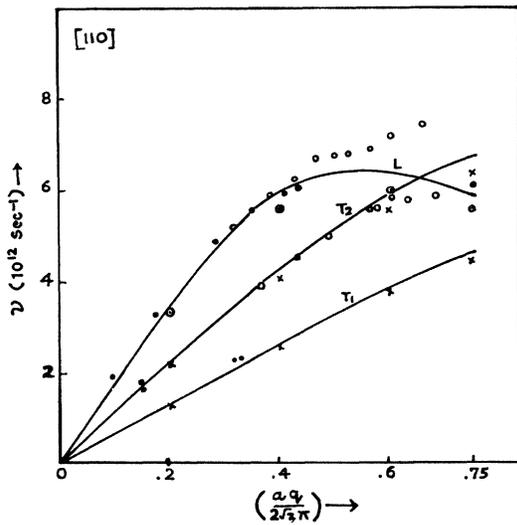


FIG. 3. Dispersion curve for copper for [110] direction. —, authors' theoretical; ○, Toya's longitudinal; ×, Toya's transverse; ○, experimental (Cribier *et al.*); ●, experimental (Sinha and Squires).

icients: $V(r_s)=0$; $E_0=-3.7$ eV; $W_F=6.95$ eV; $r_s=2.68a_0$. Some of these are very uncertain. We have, however, used the same values in the present calculations. p and q were determined by making a good fit of (14) with the true energy expression. This is shown in Fig. 1. Their numerical values are found to be

$$p=1.246, \quad q=0.022.$$

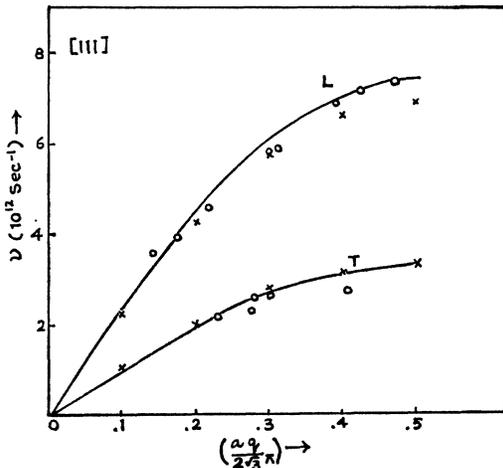


FIG. 4. Dispersion curve for copper for [111] direction. —, author's theoretical; ×, Toya's theoretical; ○, experimental (Cribier *et al.*).

Using these values for p and q , the quantity $G(u, h)$ was calculated for a few values of u from Eq. (A22) of Bailyn and a graph was plotted of $G(u, h)$ versus u . From this graph $G(u, h)$ were read for the values of u involved in the computation. $F(u)$ were calculated from Eq. (12).

The calculated frequencies in the three symmetry directions have been plotted in Figs. 2, 3, and 4 where they have been compared with the neutron-diffraction results of Cribier *et al.*^{4,28} and Sinha and Squires.⁵ The frequencies calculated by Toya have also been shown for comparison. These clearly show that our calculated

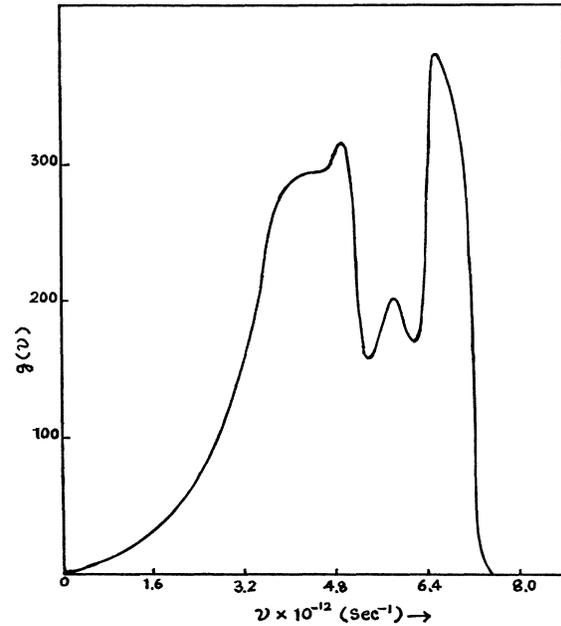


FIG. 5. Frequency distribution curve for copper.

dispersion curves are in better agreement with the neutron diffraction results than those obtained by the use of Toya's total effective matrix element. The theoretical results shown in Fig. 3 disagree with the experimental results of Cribier *et al.*⁴ in respect of crossing of the longitudinal and transverse branches in the [110]

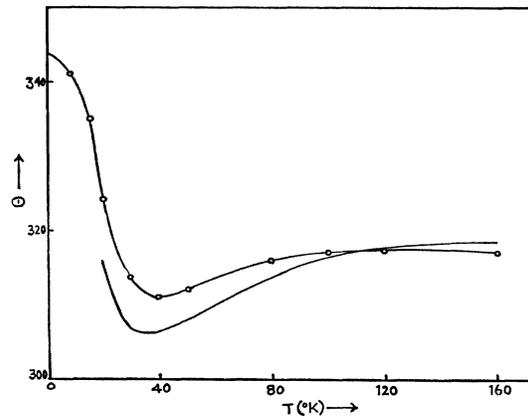


FIG. 6. θ - T curve for copper. —, theoretical; —○—, experimental (Martin).

²⁸ The experimental results of Cribier *et al.* shown here give the revised measurements communicated to the authors privately.

TABLE I. Specific heats of copper in cal mole⁻¹deg⁻¹.

Temperature °K	C_v calculated	C_v observed Martin ^a	$C_v - \gamma T$ observed
20	0.118	0.113	0.110
30	0.423	0.404	0.399
40	0.916	0.892	0.886
50	1.497	1.465	1.459
80	3.070	3.061	3.050
100	3.800	3.802	3.789
120	4.302	4.317	4.297
160	4.930	4.963	4.937

^a See Refs. 30 and 31.

direction. It appears to us that the experimental results in this region are not reliable. The theoretical calculations by other models^{5-7,20,21} and the latest experimental data of Dr. Sinha obtained in Cambridge do show this crossing.²⁹

The frequency distribution curve $g(\nu)$ is shown in Fig. 5. The curve is not normalized and the units of $g(\nu)$ are arbitrary.

 TABLE II. Calculated and experimental values of the elastic constants of copper in units of 10¹² dynes/cm².

	C_{11}	C_{12}	C_{44}	$(C_{11} - C_{12})$
Theoretical (ours)	1.54	1.06	0.79	0.48
Theoretical (Toya)	1.40	0.94	0.82	0.46
Experimental (room temp.) (Overton and Gaffney) ^a	1.69	1.22	0.76	0.47

^a See Ref. 32.

²⁹ The authors are indebted to Dr. Sinha for showing us his unpublished curves. The agreement with his neutron scattering results is very good.

The specific heats have been calculated by numerical computation using Blackman's sampling technique. For this purpose frequencies have been divided into intervals of $\Delta\nu = 0.4 \times 10^{12}$ sec⁻¹. The specific heats have been computed from Einstein functions corresponding to the midpoint of each interval. Since the mesh of points in the Brillouin zone is too coarse for getting results below 20°K, the calculations have not been extended below this temperature. The results are shown in Table I where they have been compared with the latest experimental data of Martin.^{30,31} The theoretical and experimental θ - T curves have been shown in Fig. 6. The deviation is less than 3%. The excellent agreement both with the neutron-diffraction data as well as specific heats clearly shows that the use of Baily's matrix element gives more satisfactory results.

The elastic constants have been calculated from the phonon dispersion curves graphically and are given in Table II. These have been compared with the values given by Overton and Gaffney³² for 290°K. While the calculated shear constants ($C_{11} - C_{12}$) and C_{44} show good agreement with the experimental data, there is some discrepancy in the case of C_{11} and C_{12} . In view of the uncertain values of the several parameters an overall good agreement is hardly to be expected. Nevertheless there is an obvious improvement over Toya's theoretical values which also have been shown in Table II.

The results presented in this paper form part of a thesis submitted by one of us (P. L. S.) for the Ph.D. degree of the Banaras Hindu University. P. L. S. is thankful to the authorities of the Bhagalpur University for encouragement and for leave to work at the Banaras Hindu University.

³⁰ D. L. Martin, Can J. Phys. **38**, 17 (1960).

³¹ D. L. Martin, Can J. Phys. **40**, 1166 (1962).

³² W. C. Overton and J. Gaffney, Phys. Rev. **98**, 969 (1955).