LATTICE DYNAMICAL STUDY OF COPPER

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A study of the lattice dynamics of copper has been made using Bhatia and Horton's model of electron lattice interaction. The dispersion relations obtained for the three symmetry directions have been compared with recent neutron scattering results. The complete vibration spectra and Debye temperatures have been evaluated. The theoretical results are found to be in good agreement with experiment.

1. Introduction

The lattice vibrations of metallic crystals have been the subject of considerable experimental and theoretical studies in literature. Early authors completely ignored the effect of electrons on lattice vibrations in metals. During the last few years a number of models [1–4] have been proposed where the electron gas is considered and taken into account in various approximate ways. Bhatia and Horton [2], taking into account in a simple way the effect of valence electrons on the ionic motion in monovalent cubic metals, but considering only the interaction between the nearest neighbours, have obtained satisfactory results on frequency spectra and lattice specific heats for sodium and silver. This model was further found to show fairly well the gross features of the frequency distributions and to give satisfactory values of the heat capacities for alkali metals and gold (Joshi and Hemkar [5]). Recently Sangal and Sharma [6] have used this model to investigate the temperature variation of electrical and thermal resistivities of alkali metals and thermal expansion of a number of cubic metals with satisfactory results.

In the present paper, we apply the Bhatia model to the calculations of the vibrational spectrum and specific heat of copper. The choice of the substance is dictated by the fact that more reliable inelastic neutron scattering results on phonon dispersion are now available. The study provides a further check of the accuracy of the model.

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2. Theory

The secular equation determining the angular frequencies ω of the elastic waves is written as

$$\lambda\varrho\omega^2 = \sum_{l,m,n}\left\{\frac{4}{a^2}\left[\sin^2\tfrac{1}{2}ka(Ll+Mm+Nn)\right]\times\left[\tfrac{1}{4}(C_{44}+\varepsilon)\lambda-\varepsilon l(l\lambda+m\mu+n\nu)\right]\right\}+$$

$$+\frac{(C_{11}-C_{44}+\varepsilon)L(\lambda L+\mu M+\nu N)k^2}{1+(C_{11}-C_{44}+\varepsilon)\,(k^2/4\pi e^2 n_0^2)} \tag{1a}$$

with two similar equations obtained by putting

$$l \rightleftarrows m,\ y \rightleftarrows \mu, \text{ an } L \rightleftarrows M \tag{1b}$$

and

$$l \rightleftarrows n,\ \lambda \rightleftarrows \nu, \text{ and } L \rightleftarrows N. \tag{1c}$$

Here (L, M, N), (λ, μ, ν) and (l, m, n) are the direction cosines of the propagation vector k, the displacement vector and the line joining the ion at the origin and a nearest neighbour ion, respectively. C_{11}, C_{12} and C_{44} are the elastic constants, $\varepsilon = C_{11}-C_{12}-2C_{44}$, ϱ the density, n_0 the number of electrons per cm^3, a the nearest neighbour distance and the summation in (1) is over all the nearest neighbours.

The secular equation has been solved for k lying along the three directions $A(100)$, $B(110)$ and $C(111)$ and the solutions are as follows:

$A(100)$: Longitudinal branch

$$\omega^2 = \left(\frac{1}{a^2\varrho}\right)8(C_{44}-\varepsilon)\sin^2\left(\frac{ka}{2\sqrt{2}}\right)+\frac{(C_{11}-C_{44}+\varepsilon)k^2}{\varrho(1+\chi k^2a^2)}. \tag{2a}$$

Transverse branch

$$\omega^2 = \left(\frac{1}{a^2\varrho}\right)8C_{44}\sin^2\left(\frac{ka}{2\sqrt{2}}\right). \tag{2b}$$

$B(110)$: Longitudinal branch

$$\omega^2 = \left(\frac{1}{a^2\varrho}\right)\left[2(C_{44}-3\varepsilon)\sin^2\left(\frac{ka}{2}\right)+8C_{44}\sin^2\left(\frac{ka}{4}\right)\right]+$$

$$+\frac{(C_{11}-C_{44}+\varepsilon)k^2}{\varrho(1+\chi k^2a^2)}. \tag{2c}$$

Transverse branch

$$\omega^2 = \left(\frac{1}{a^2\varrho}\right)\left[2(C_{44}+\varepsilon)\sin^2\left(\frac{ka}{2}\right)+8C_{44}\sin^2\left(\frac{ka}{4}\right)\right]. \tag{2d}$$

Transverse branch

$$\omega^2 = \left(\frac{1}{a^2\varrho}\right)\left[2(C_{44}+\varepsilon)\sin^2\left(\frac{ka}{2}\right)+8(C_{44}-\varepsilon)\sin^2\left(\frac{ka}{4}\right)\right]. \tag{2e}$$

$C(111)$: Longitudinal branch

$$\omega^2 = \left(\frac{1}{a^2\varrho}\right)2(3C_{44}-5\varepsilon)\sin^2\left(\frac{ka}{\sqrt{6}}\right)+\frac{(C_{11}-C_{44}+\varepsilon)k^2}{\varrho(1+k^2a^2)}. \tag{2f}$$

Transverse branch

$$\omega^2 = \left(\frac{1}{a^2\varrho}\right)2(3C_{44}+\varepsilon)\sin^2\left(\frac{ka}{\sqrt{6}}\right), \tag{2g}$$

where

$$\chi a^2 = (C_{11}-C_{44}+\varepsilon)/4\pi e^2 n_0^2.$$

In Houston's approximation, the normalized frequency distribution function $G(\omega)$ is given by

$$G(\omega) = \frac{FV}{(2\pi)^3}\cdot\frac{4\pi}{35}\sum_i\left[10k^2\left(\frac{dk}{d\omega}\right)_{i,A}+16k^2\left(\frac{dk}{d\omega}\right)_{i,B}+9k^2\left(\frac{dk}{d\omega}\right)_{i,C}\right]$$

where V is the volume of the crystal, F the normalization factor and the summation is over the three polarization branches for each k.

The gram atomic specific heat C_v is obtained in the usual manner by numerical integration from the frequency spectrum of Fig. 3.

3. Results and discussion

The elastic constants used in the calculation are taken from the measurements of Svensson *et al.* [9]. These and other constants needed in the calculation are listed below:

Elastic constants	At 0°K$_{\text{dyne/cm}^2}$	At 296°K$_{\text{dyne/cm}^2}$
C_{11}	17.62×10^{11}	16.85×10^{11}
C_{12}	12.49×10^{11}	12.15×10^{11}
C_{44}	8.18×10^{11}	7.55×10^{11}

$a = 2.546$ Å and $\varrho = 8.94$ gm/c.c.

The calculated dispersion curves of copper together with the experimental points are shown in Fig. 1. The experimental inelastic neutron scattering results on phonon dispersion relations in copper are due to various groups of authors [7–9]. We have compared our theoretical results with those of Svensson *et al.* in the three symmetry directions and have obtained fairly good agreement.

The frequency distributions $G(\omega)$ corresponding to elastic constants at 296°K and 0°K are plotted against ω in Figs 2 and 3. As usual the singularities of $G(\omega)$, characteristic of Houston's approximation, have been replaced by finite peaks of equal area. For com-

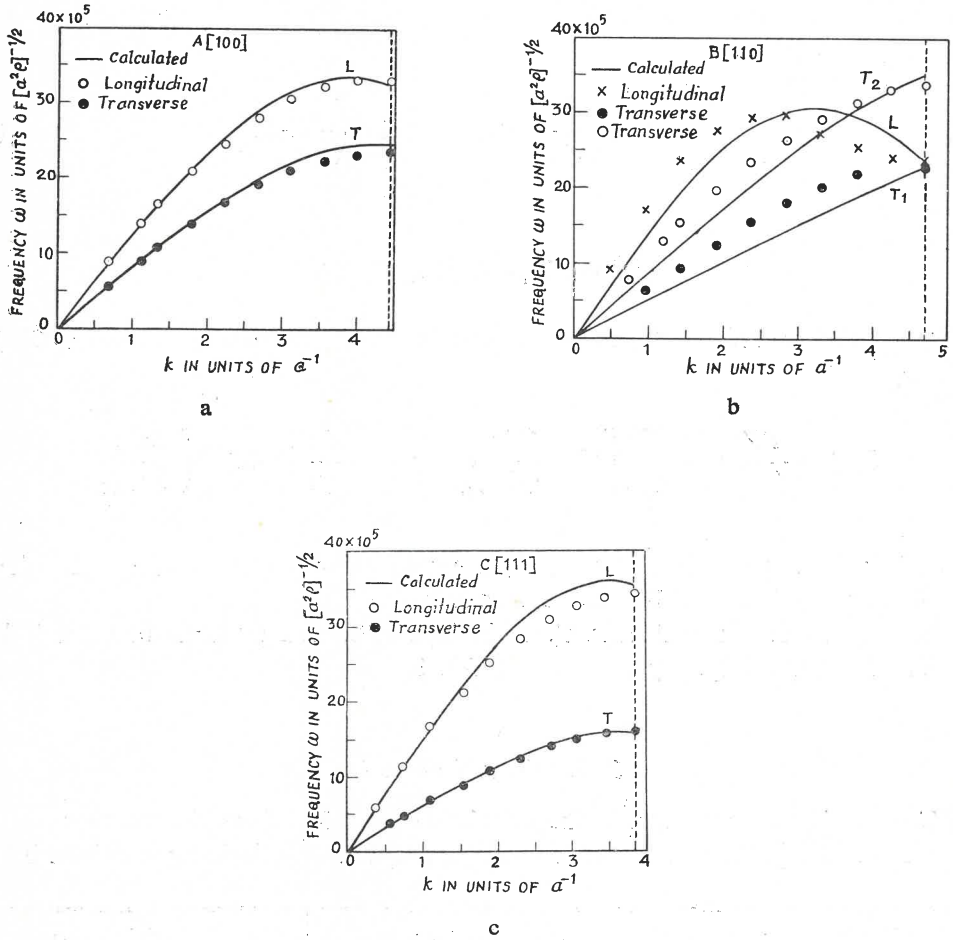

Fig. 1. Phonon dispersion curves at room temperature in the three symmetry directions in copper. The experimental results are due to Svensson *et al.*

parison the distribution curves of Jacobsen [10] and Svensson *et al.* are also plotted in Fig. 2. Jacobsen has calculated the frequency spectrum of copper from the diffuse X-ray scattering measurements at room temperature. The calculated Debye characteristic temper-

atures Θ are shown in Fig. 4, along with the experimental points due to Martin [11, 12]. As it is obvious from the figure, satisfactory agreement between the theoretical and experimental results is obtained.

Thus we conclude that the Bhatia model offers a fairly resonable explanation for the thermal properties of copper. The small discrepancies between the theory and experiment are not unexpected and may be attributed to: 1) the approximate method of calcula-

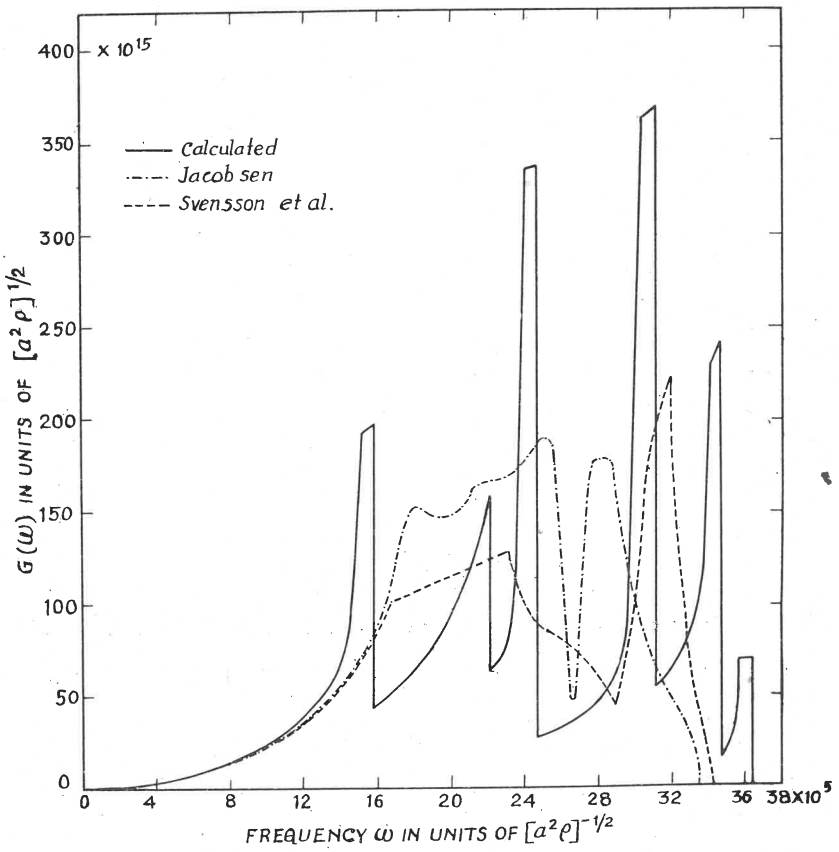

Fig. 2. Frequency spectrum of copper obtained from the elastic data at room temperature. The solid line shows present calculations, broken line corresponds to the experimental neutron scattering results of Svensson *et al.* and dash-dotted line represents Jacobsen's calculations from *X*-ray measurements at room temperature

ting the frequency spectrum, 2) the neglect of anharmonic effects at higher temperatures and 3) the neglect of the temperature variation of elastic constants.

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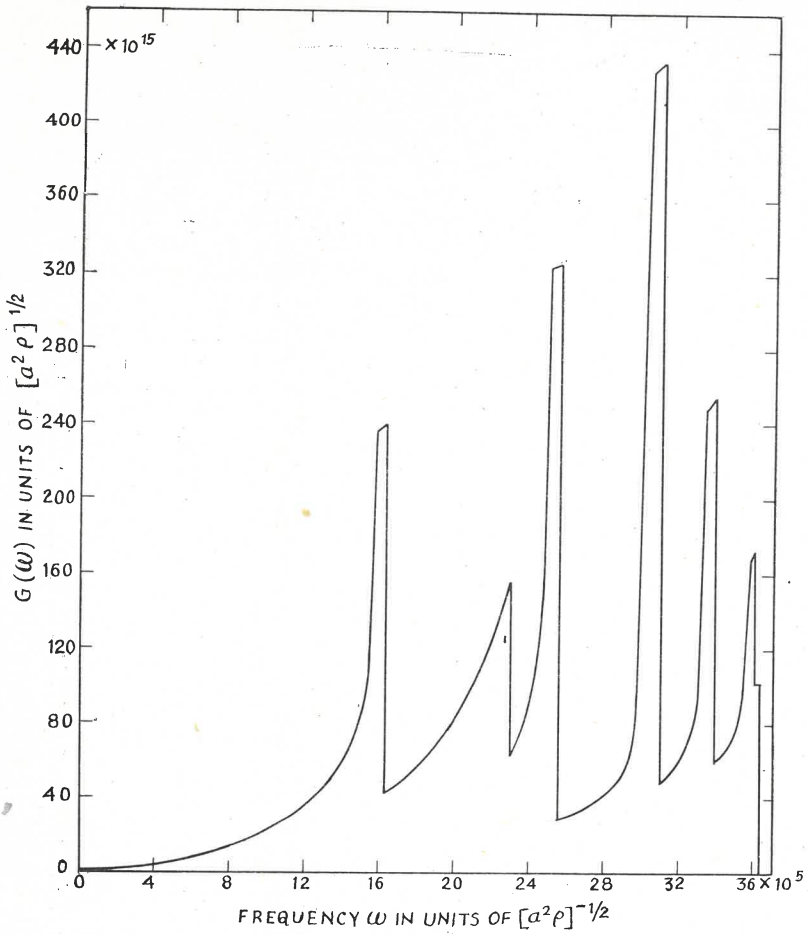

Fig. 3. Frequency spectrum of copper obtained from the elastic data at 0°K

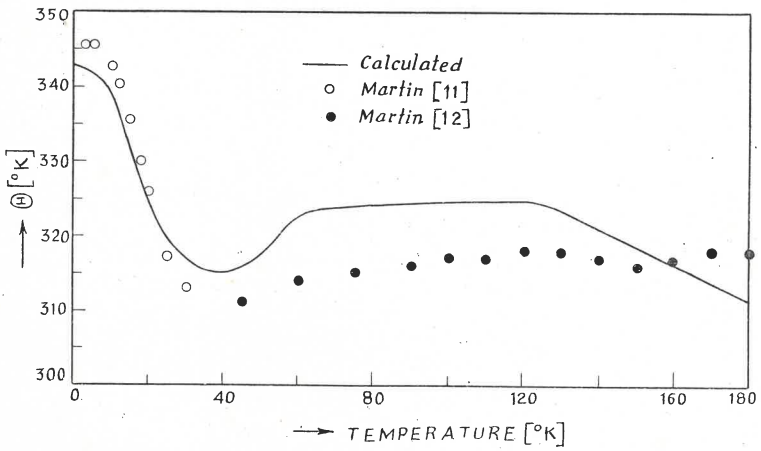

Fig. 4. The Debye characteristic temperature as a function of temperature. The solid curve is calculated from the frequency spectrum shown in Fig. 3. The experimental points are due to Martin

REFERENCES

[1] J. de Launay, *J. Chem. Phys.*, **21**, 1975 (1954).
[2] A. B. Bhatia, *Phys. Rev.*, **97**, 363 (1955); A. B. Bhatia, G. K. Horton, *Phys. Rev.*, **98**, 1715 (1955).
[3] P. K. Sharma, S. K. Joshi, *J. Chem. Phys.*, **39**, 2633 (1963).
[4] K. Krebs, *Phys. Letters*, **10**, 12 (1964).
[5] S. K. Joshi, M. P. Hemkar, *J. Chem. Phys.*, **34**, 1458 (1961); *Phys. Rev.*, **122**, 13 (1961); *Physica*, **27**, 793 (1961); M. P. Hemkar, S. K. Joshi, *J. Phys. Soc. Japan*, **17**, 754 (1962).
[6] S. K. Sangal, P. K. Sharma, *Acta Phys. Chem.*, **15**, 35 (1969); *Czech. J. Phys.*, Bi **19**, 1095 (1969).
[7] S. K. Sinha, *Phys. Rev.*, **143**, 422 (1966).
[8] R. M. Nicklow, G. Gilat, H. G. Smith, L. J. Raubenheimer, M. K. Wilkinson, *Phys. Rev.*, **164**, 922 (1967).
[9] E. C. Svensson, B. N. Brockhouse, J. M. Rowe, *Phys. Rev.*, **155**, 619 (1967).
[10] E. H. Jacobsen, *Phys. Rev.*, **97**, 654 (1955).
[11] D. L. Martin, *Phys. Rev.*, **141**, 576 (1966).
[12] D. L. Martin, *Canad. J. Phys.*, **38**, 17 (1960).