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Lattice dynamical study of noble metals (I)

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Abstract. The lattice dynamical study of noble metals (Cu, Ag and Au) has been made on the basis of improved Clark–Gazis–Wallis model considering volume forces of Krebs’ nature. The frequency/wave-vector dispersion relations obtained for the three symmetry directions have been compared with recent inelastic neutron scattering experiments. The computation of the specific heats at constant volume have been made by Blackman’s root sampling technique for temperatures above $\Theta/10$ and below this temperature the calculations are carried out by employing the modified Houston method. The computed lattice specific heats and the effective Debye temperatures are compared with the available calorimetric data. The theory is in good agreement with the experimental data.

1. Introduction

The presence of conduction electrons in metallic crystals introduces complicating features as it essentially involves the study of a system with many degrees of freedom quite difficult to handle and a rigorous solution of the problem becomes a difficult task. The presence of these electrons led the early workers to completely ignore their effect on the lattice vibrations, therefore their approaches represented an incomplete picture of the total interactions taking place in cubic crystals.

However, in the recent past a number of models [1–3] have been proposed to study the lattice vibrations in metals taking also into account the presence of conduction electrons. But these approaches could not stand the test of the experiments due to the fact that the frequencies obtained from them are not periodic in reciprocal space due to the neglect of translational invariance. It seems, therefore, worthwhile to consider a more realistic model free of the above defect for the lattice dynamical description of cubic metals.

Quite recently, Shukla et al. [4] suggested a modification in the Clark–Gazis–Wallis (CGW) angular force model [5] for fcc metals by incorporating the effect of the presence of conduction electrons, an aspect of interaction which is very important for metals. It was also emphasized that the Cauchy discrepancy in metals arises from the volume forces in addition to the central and angular interactions of the usual CGW nature. This improvement of the original CGW approach seems to be essential because the presence of electron gas forms an integral part of lattice interactions and should be adequately taken into consideration in a lattice dynamical approach. The inclusion of the Umklapp process by Shukla et al. in the modified CGW angular force model, by the consideration of volume forces of Krebs [6]

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nature, enables it to satisfy the symmetry requirements of the lattice. This modified force model considers central and angular interactions up to second neighbours only and has been used successfully by them for the phonon dispersion study of fcc metal copper. A similar improvement in the CGW angular force model for bcc metals has also been made recently by the present authors [7] and is used for the dispersion study of a number of bcc metals [7, 8–11]. In view of nearness of such an approach to the actual physical interactions taking place in metals and the success with which it interprets the phonon dispersion of both fcc and bcc metals [7, 8–11], it was thought suitable to test its usefulness in studying the lattice dynamics of copper, silver and gold, which fall in the group of noble metals. The present paper reports the phonon dispersion, specific heats at constant volume and the equivalent Debye temperature of these metals and these calculations provide a further check on the validity of the modified CGW model for lattice dynamical study of cubic metals.

2. Secular determinant

The phonon frequencies for lattice vibration can be obtained from the secular equation

$$|D(\mathbf{q}) - 4m\pi^2 v^2 \mathbf{I}| = 0 \quad (1)$$

where \mathbf{D} is 3×3 dynamical matrix, \mathbf{I} the unit matrix and m the ionic mass. The explicit expression for typical elements of \mathbf{D} in the fcc structure can be given by

$$D_{11}(\mathbf{q}) = 2 \left[\alpha_1 + \frac{8}{a^2} (\gamma_1 + \gamma_2) \right] \left[2 - C_1(C_2 + C_3) \right] + 4\beta_1 S_1^2 - \frac{4\gamma_1}{a^2} (2 \cos 2\pi a k_1 - \cos 2\pi a k_2 - \cos 2\pi a k_3) + A \sum_{\mathbf{G}} \left[\frac{(\mathbf{q} + \mathbf{G})_1 (\mathbf{q} + \mathbf{G})_1}{|\mathbf{q} + \mathbf{G}|^2 + k_c^2} g^2(|\mathbf{q} + \mathbf{G}|r_s) - \frac{G_1^2}{|\mathbf{G}|^2 + k_c^2} g^2(|\mathbf{G}|r_s) \right] \quad (2)$$

$$D_{12} = \left(2\alpha_1 - \frac{16\gamma_1}{a^2} \right) S_1 S_2 + A \sum_{\mathbf{G}} \left[\frac{(\mathbf{q} + \mathbf{G})_1 (\mathbf{q} + \mathbf{G})_2}{|\mathbf{q} + \mathbf{G}|^2 + k_c^2} g^2(|\mathbf{q} + \mathbf{G}|r_s) - \frac{\mathbf{G}_1 \mathbf{G}_2}{|\mathbf{G}|^2 + k_c^2} g^2(|\mathbf{G}|r_s) \right] \quad (3)$$

where $C_1 = \cos \pi a k_1$, $S_1 = \sin \pi a k_1$, $g(x) = 3(\sin x - x \cos x)/x^3$, k_1, k_2, k_3 are components of \mathbf{k} in Cartesian coordinates, and a is the lattice constant. The factors $k_c, g(|\mathbf{G}|r_s)$ are the same as in Krebs model [6], \mathbf{G} the reciprocal lattice vector, α_1 and β_1 are the central force constants and γ_1 and γ_2 are the angular force constants up to second neighbours, the $g(x)$ function introduced above is to arbitrarily reduce the effect of larger \mathbf{G} values [12, 13]. The screening parameters k_c are given by

$$k_c^2 = k_c^2(P) f(t)$$

where $k_c(P)$ is the screening parameter of Pines [14] and is written as

$$k_c^2(P) = 0.353 \left(\frac{r_s}{a_0} \right)^{1/2} k_F.$$

Here $r_s = (3/4\pi n_e)^{1/3}$ = interelectronic spacing, n_e the electron density, a_0 is Bohr radius and $f(t)$ is given as

$$f(t) = \frac{1}{2} + \frac{1-t^2}{4t} \ln \frac{1+t}{1-t} \text{ with } t = k/2k_F.$$

In the longwavelength limit (e.g., for $k \rightarrow 0$) the force constants involved in the dynamical matrix of fcc lattice given in equations (2) and (3), can be related to the measured elastic constants and two zone boundary frequencies by the following expressions

$$\begin{aligned} aC_{11} &= 2\alpha_1 + 4\beta_1 + 32 \frac{\gamma_1}{a^2} + 16 \frac{\gamma_2}{a^2} + \frac{4A}{a^2 k_c^2} \\ aC_{12} &= \alpha_1 - \frac{16\gamma_1}{a^2} - \frac{8\gamma_2}{a^2} + \frac{4A}{a^2 k_c^2} \\ aC_{44} &= \alpha_1 + \frac{8\gamma_2}{a^2} \\ \pi^2 m v_T^2 &= \alpha_1 + 8 \frac{\gamma_1}{a^2} + 8 \frac{\gamma_2}{a^2} \\ \pi^2 m v_L^2 &= 2\alpha_1 + \beta_1 + \frac{8\gamma_2}{a^2} + AC \end{aligned} \quad (4)$$

where v_T and v_L correspond to the transverse and longitudinal zone boundary frequencies in $[\zeta 00]$ and $[\zeta \zeta \zeta]$ directions and C is the electronic contribution at the zone boundary in $[\zeta \zeta \zeta]$ direction. This set of equations contain five unknown constants (viz. α_1 , β_1 , γ_1 , γ_2 and A). Solution of these equations determine the unknown force constants in terms of elastic constants and two known zone boundary frequencies. The resulting expressions are

$$\begin{aligned} \frac{\gamma_1}{a^2} &= \frac{1}{32} [m\omega_T^2 - 4aC_{44}] \\ A &= a^2 k_c^2 / (12 - a^2 k_c^2 C) [aC_{11} + 2aC_{12} + 4aC_{44} - m\omega_L^2] \\ \frac{\gamma_2}{a^2} &= \frac{1}{16} \left[\frac{4A}{a^2 k_c^2} - aC_{12} + aC_{44} - 16\gamma_1/a^2 \right] \\ \alpha_1 &= aC_{44} - 8\gamma_2/a^2 \\ \beta_1 &= \frac{1}{4} \left[m\omega_L^2 - 8\alpha_1 - 32 \frac{\gamma_2}{a^2} - AC \right]. \end{aligned} \quad (5)$$

These force constants are substituted to find the roots (frequencies) of the secular determinant for selected values of phonon wave vectors.

3. Numerical computation

The frequency versus wave vector dispersion relations at room temperature along the three symmetry directions $[\zeta 00]$, $[\zeta \zeta 0]$ and $[\zeta \zeta \zeta]$ of silver and gold are

determined from the solutions of secular equation (1) along these directions. The dispersion curves of copper have already been reported [4].

The elastic constants of these metals at room temperature and other relevant parameters used in the calculations are given in Table 1. The phonon dispersion

Table 1
Physical constants for metals used in the calculations

Metal	Lattice constant (Å)	Elastic constants			References
		C_{11}	C_{12}	C_{44}	
Copper	3.603	17.62	12.49	8.18	[a, b]
Silver	4.060	13.15	9.73	5.11	[c, a]
Gold	4.056	20.16	16.97	4.54	[a]

[a] C. Kittel, In *Introduction to Solid State Physics* (3rd Edn.) 29, 122 (1967).

[b] E. C. Svensson, B. N. Brockhouse and J. M. Rowe, *Phys. Rev.* 155, 619 (1967).

[c] W. A. Kamitakahara and B. N. Brockhouse, *Phys. Lett.* 29A, 639 (1969).

curves, together with the experimental data of a recent neutron scattering experiment, are plotted in Figures 1–2. The frequency distribution of the normal modes of vibrations have been computed by Blackman's root sampling technique for a discrete subdivision in wave-vector space. For this purpose it is essential to determine the characteristic frequencies at a suitable number of points in an irreducible section of the first Brillouin zone. We have considered an evenly distributed mesh of 8000 wave vectors in the first Brillouin zone. From Born's cyclic boundary condition and considerations of lattice symmetry, these 8000 points reduce to 262 non-equivalent points, including the origin lying within $\frac{1}{48}$ th part of the Brillouin zone and weighted according to the number of similar points associated with them. The 24 000 frequencies corresponding to 8000 points in the zone were obtained from the solution of the secular equation of the modified CGW model for an fcc lattice. These frequencies represent the complete vibration spectrum. The number of frequencies lying in the intervals of 0.14×10^{12} , 0.12×10^{12} , 0.9×10^{12} (all in c/s) for copper, silver and gold were counted and from these the frequency distribution curves of the respective metals were obtained. Using the computed frequency spectra of the metals, the contribution of lattice vibrations to the specific heat per gram atom i.e. C_v was obtained from the numerical integration of the usual relation

$$C_v = 3R \int_0^{\omega_m} E(\hbar\omega/kT)G(\omega) d\omega \quad (6)$$

where R is the gas constant, T the absolute temperature, ω_m the maximum frequency and $E(\hbar\omega/kT)$ the Einstein specific heat function. This method was used at moderate temperatures down to $\frac{1}{10}\Theta$ as $G(\omega)$ calculated by the sampling method cannot be very accurate in the low-frequency range and because of the coarseness of the mesh used. For temperatures below $\frac{1}{10}\Theta$, where the lower frequencies have a predominating influence, we used Houston's method [15] as elaborated by Horton and Schiff [16]. The frequency distribution function can be written as

$$G(\omega) = \left(\frac{1}{2\pi}\right)^3 \int \sum_p \left(q^2 \frac{dq}{d\omega}\right)_p d\Omega_q, \quad (7)$$

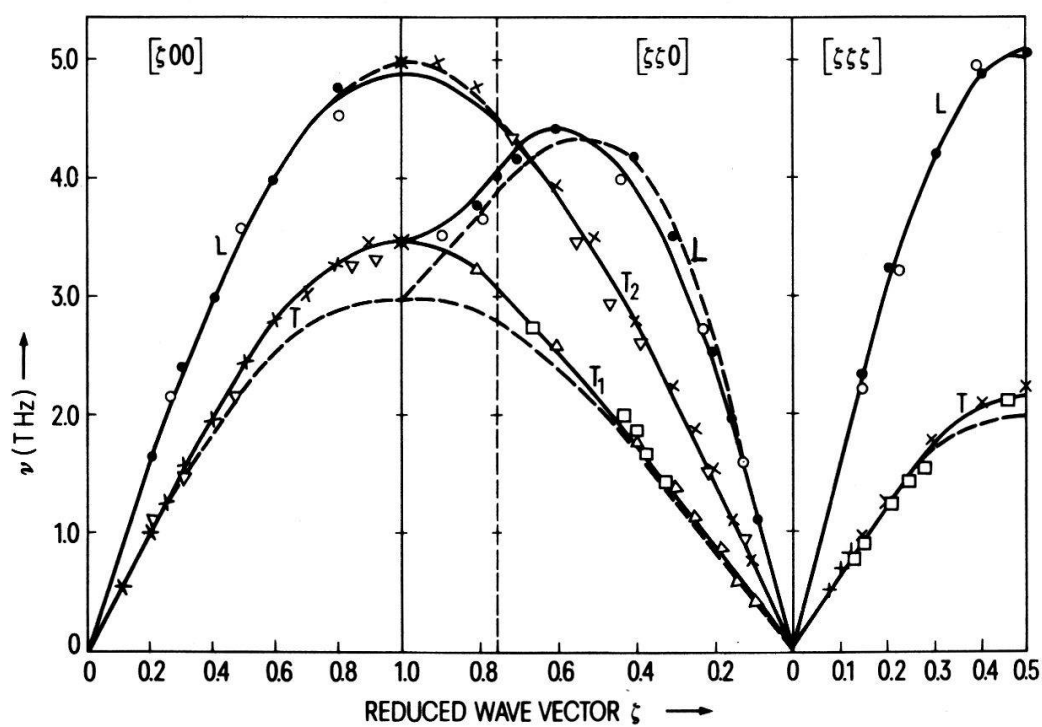


Figure 1
Dispersion curves along the symmetry directions for silver.

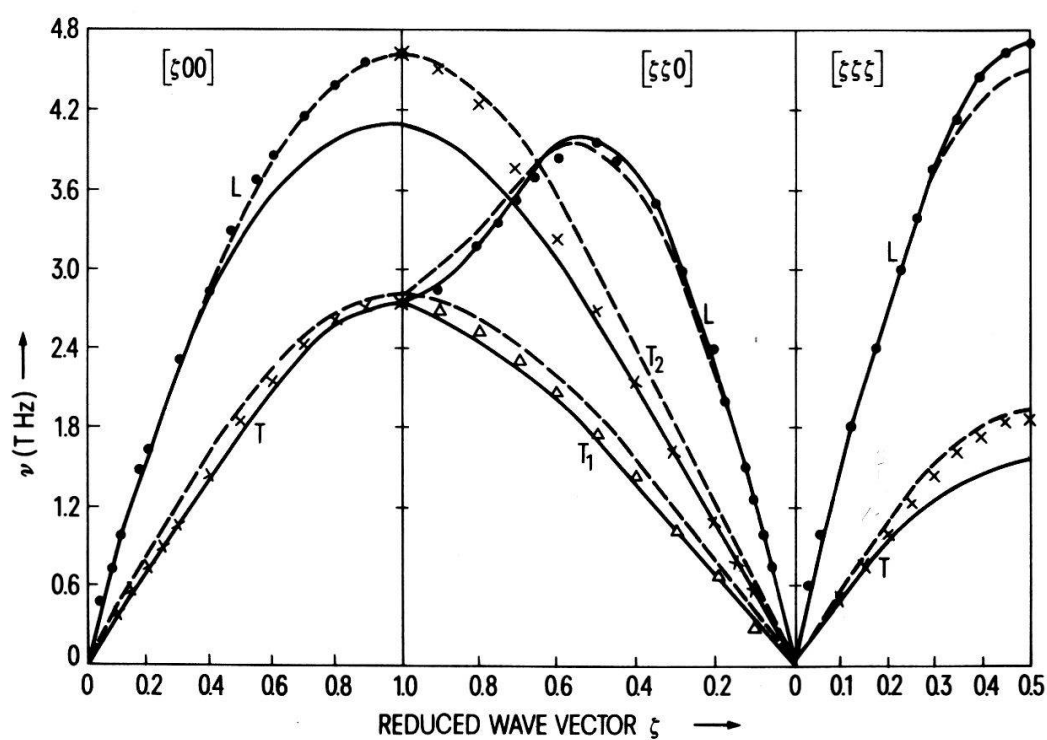


Figure 2
Dispersion curves along the symmetry directions for gold.

where Ω_q is the solid angle in the wave vector space and the summation over p is on the three modes of lattice vibrations for each \mathbf{q} . Use of (6) in (7) gives

$$C_v = \frac{3R}{(2\pi)^3} \sum_p \int_0^{q_{\max}} \int_{\Omega_q} E\left(\frac{\hbar\omega_p}{kT}\right) q^2 dq d\Omega. \quad (8)$$

For the evaluation of C_v from (8), the integration over \mathbf{q} was performed numerically and the angular integration was carried out by a six-term interpolation procedure.

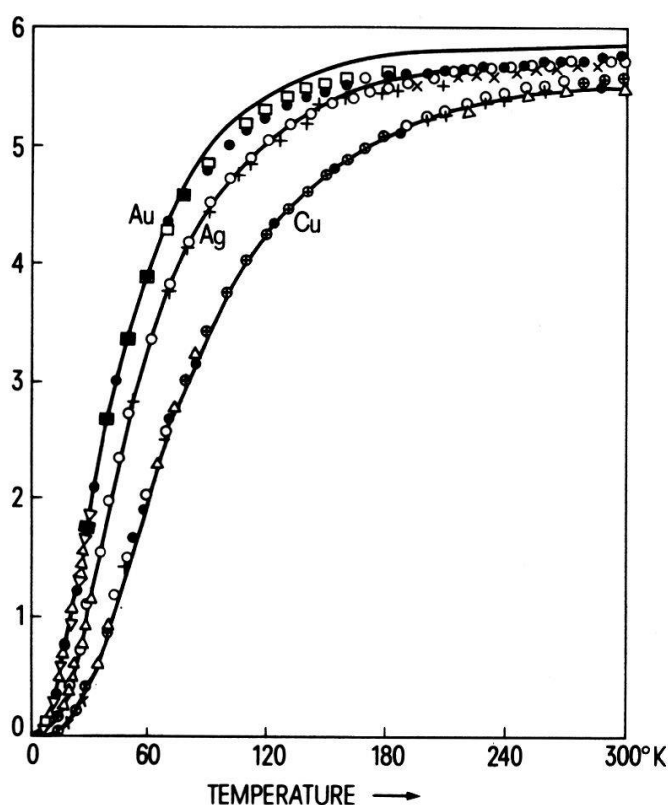


Figure 3
The lattice specific heats of copper, silver and gold along with experimental values.

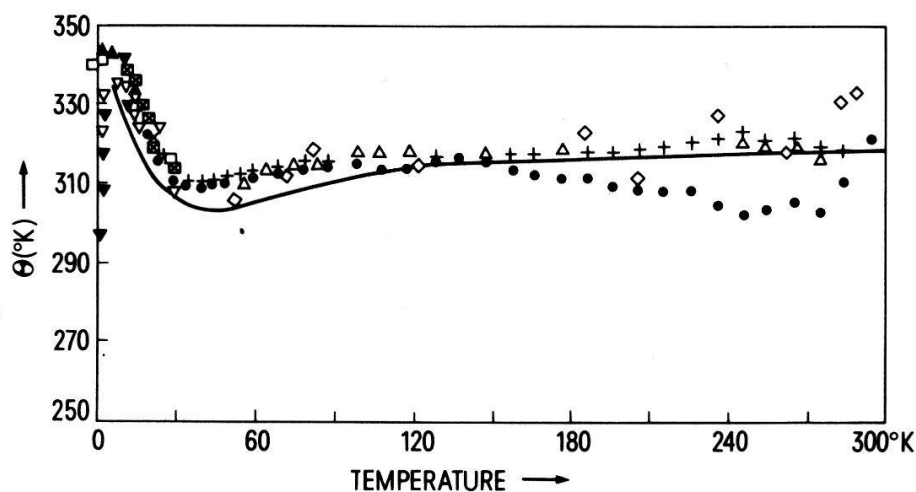


Figure 4
The Debye characteristic temperature of copper as a function of temperature.

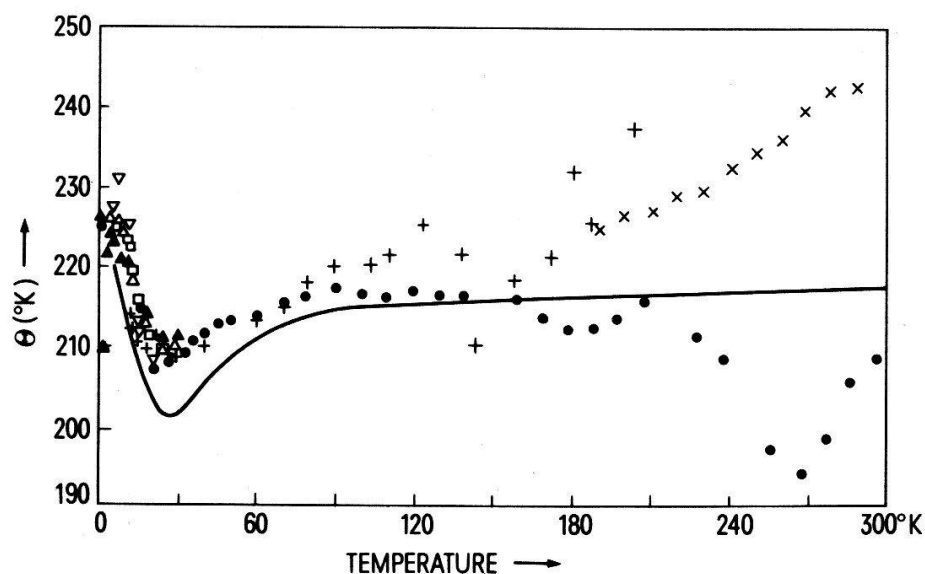


Figure 5
The Debye characteristic temperature of silver as a function of temperature.

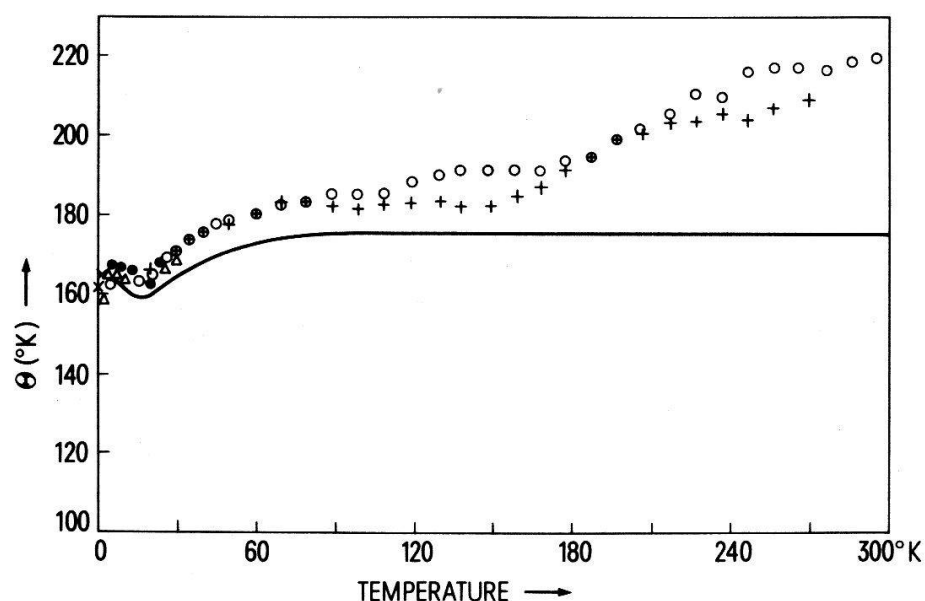


Figure 6
The Debye characteristic temperature of gold as a function of temperature.

The six directions of \mathbf{q} used are: $[100]$, $[110]$, $[111]$, $[210]$, $[211]$ and $[221]$. The calculated results together with the observed C_v (corrected for $C_p - C_v$ and electronic specific heat) for the three noble metals at various temperatures are plotted in Figure 3. Another comparison with the experimental data on C_v is made in terms of the equivalent Debye temperature Θ and is depicted in Figures 4–6.

4. Results and discussion

4.1. Dispersion relations

The room temperature dispersion relations of silver have been recently reported by Drexel et al. [17] and Kamitakahara and Brockhouse [18], the later results being

more extensive. However, there is considerable discrepancy between the results of both the groups at higher frequencies. The experimental points of both have been plotted in Figure 1. Quite recently, Lynn et al. [19] have reported the measurements of phonon dispersion relations at room temperature in the higher symmetry directions of gold using a coherent inelastic scattering of neutrons. The experimental values of these observations are plotted in Figure 2. In Figures 1–2 solid lines represent the present calculations and are in fairly satisfactory agreement with the experimental data, except for some deviations at the zone boundary along some directions. This discrepancy may be attributed to the assumption of short range interionic forces in the secular equation.

Further, the frequency/wave vector dispersion curves of silver and gold have also been compared with the recently published phonon frequencies of Singh and Prakash [20], calculated from the Animalu's [21] transition metal model potential (TMMP) which includes s – d hybridization and is used in conjunction with the dielectric function. The computed curves of these authors have been indicated by dotted lines on Figures 1 and 2. Except for the longitudinal and T_2 branches of the $[\zeta 00]$ and $[\zeta \zeta 0]$ directions of gold, the results obtained with our model exhibit a comparatively better agreement with the experiment. The results obtained by Singh and Prakash, especially in the $[\zeta 00]$ and $[\zeta \zeta 0]$ symmetry directions, show a poor agreement with the experimental observations.

4.2. Specific heat

The specific heat at constant volume C_v per gram atom of noble metals has been calculated in the usual manner by numerical integration. In the present case the calculation for each temperature was made separately using the input data appropriate to that temperature. The experimental values of the temperature variation of the elastic constants for copper were obtained from the measurements of Overton and Gaffney [22], while for silver and gold these were taken from Neighbours and Alers [23]. The lattice parameter values at various temperatures were taken from Pearson [24]. The experimental data on heat capacity of copper are too numerous to be described in detail. Earlier measurements in the temperature range 20–300°K are on relatively impure samples and are not suitable for the estimation of a reliable Debye temperature. Most of them exhibit scatter due to impurities in the sample. Eucken and Werth [25], Maier and Anderson [26], Giaque and Meads [27], Dockerty [28] and Martin [29] have accurately measured the heat capacity of variously treated samples of copper in the interval 20–300°K. The measurements of Martin show good agreement with those of Dockerty, but are comparatively lower than the measurements of Giaque and Meads and he attributed this discrepancy to the perfection of the samples. For silver the only reliable data in the temperature range 15–300°K is due to Meads et al. [30]. Their results agree very well with those of Eucken et al. [31] measured in the temperature range 11 to 205°K. The accurate calorimetric data for gold in between 30–300°K have been reported by Geballe and Giaque [32], and Franzosini and Clusius [33]. A number of measurements in the low temperature range 4–20°K are available for noble metals. The earlier work carried out by Kok and Keesom [34] was on a relatively impure sample and cannot give a reliable value of the Debye temperature. The recent studies by Du Chatenier and Noble [35] show deviations of several percent from the observations of the majority of workers

Table 2
Source of experimental specific heat data and their temperature ranges

Metal	Temperature range (°K)	Source	Ref.
Copper	53 – 293	Maier and Anderson	[26]
	30 – 300	Dockerty	[28]
	20 – 300	Martin	[29]
	15 – 300	Giauque and Meads	[27]
	15 – 90	Martin	[29]
	3 – 30	Martin	[36]
	2.3 – 19.7	Ahlers	[37]
	1.3 – 30	du Chatenier and Noble	[35], [a]
	1.2 – 20	Kok and Keesom	[34]
	1 – 30	Cetas et al.	[38]
Silver	193 – 293	Broonson and Wilson	[b]
	15 – 300	Meads et al.	[30]
	11 – 205	Eucken et al.	[31]
	3 – 30	Martin	[36]
	1.4 – 26	Ahlers	[37]
	1.35 – 20.3	Keesom and Kok	[34]
	1.3 – 30	du Chatenier and Noble	[a]
	0.4 – 1.5	Filby and Martin	[c]
Gold	15 – 300	Geballe and Giauque	[32]
	12 – 273	Franzosini and Clusius	[33]
	3 – 30	Martin	[36]
	1.3 – 30	du Chatenier et al.	[35]
	0.4 – 1.5	Martin	[d]

[a] F. J. du Chatenier and J. de Noble, *Physica* 28, 181 (1962).

[b] H. L. Bronson and A. J. C. Wilson, *Canad. J. Research* 14A, 181 (1936).

[c] J. D. Filby and D. L. Martin, *Canad. J. Phys.* 40, 791 (1962).

[d] D. L. Martin, *Phys. Rev.* 170, 650 (1968).

over the whole temperature range. Martin [36], Ahlers [37], and Cetas et al. [38] have made detailed measurements of specific heat of noble metals in the interval 3–30°K. The calorimetric data for noble metals selected for present comparison are shown in Table 2. To obtain C_v from C_p , we have used the relation

$$C_p/C_v = 1 + AC_pT, \quad (9)$$

where A is a constant which can be evaluated from the well-known thermodynamic relation

$$C_p - C_v = (\alpha^2 M / JK_p) T. \quad (10)$$

Here α is the volume coefficient of thermal expansion, K the compressibility, M the atomic weight, p the density and J the mechanical equivalent of heat. The values of isothermal compressibility for copper were obtained from ultrasonic measurements of Overton and Gaffney [39] and for gold and silver these are taken from the work of Neighbours and Alers [40]. The thermal expansion data for noble metals at different temperatures were taken from several sources. For copper the data of Carr [41] et al., Rubin et al. [42], and Leksina and Novikova [43] were taken in the temperature range 6–30°K, 30–90°K and 90–300°K, respectively. For silver below

90°K the thermal expansion data of Fraser and Hollis-Hallet [44] has been considered and for temperatures above 90°K the measurements of Nix and MacNair [45] and Lekaina and Novikava [43] have been used in calculations. The thermal expansion data for gold are due to Kos et al. [46] in the temperature range 5–15°K and of Nix and MacNair [47], Leksina and Novikova [43], and Fraser and Hollis-Hallet [44] at temperatures above 15°K. Below 20°K the correction to C_p was found negligible.

The C_v values were also corrected for the electronic specific heat. The values of electronic specific heat coefficient γ used for this purpose are given in Table 3.

Table 3
Electronic specific heat coefficients for noble metals (cal/°K² g-atom)

Metal	
Copper	165.2 ^a)
Silver	153.1 ^a)
Gold	165.1 ^a)

^a) D. L. Martin, Phys. Rev. 170 (1968).

These are taken from the low temperature measurements of Martin. The C_v curves of noble metals calculated by the present model, along with the experimental data due to different workers, are plotted in Figure 3. A comparison has also been made with the experimental specific heats in terms of equivalent Debye temperatures. These Θ – T curves have been plotted in Figures 4–6. The calculated Θ – T values are in reasonably satisfactory agreement with experimental values for copper. For silver and gold the experimental values are higher than the theoretical ones and the discrepancy increases with the rise in temperature. The discrepancy between the theory and experiment in their case may be attributed to the large scatter in the experimental points at higher temperatures which is particularly striking in the case of silver. This large uncertainty in the experimental measurements therefore makes it difficult to have a comparative study of the two in the high temperature region. This discrepancy at higher temperatures may also be partly attributed to the negligence of anharmonic effects and the assumption of short-range interionic interactions in the theory. Nevertheless, the present study shows that the modified CGW approach gives a reasonably satisfactory description of lattice dynamics of noble metals.

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