Debye Waller Factors of fcc Metals by the Modified Angular Force Model

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A modified form of the non-central force model which takes account of the electron-ion interaction term of the Kreb's model in the Clark, Gazis and Wallis type angular forces is considered to calculate the Debye-Waller exponents at different temperatures for five fcc metals: copper, silver, gold, aluminium and nickel. The results are compared with the available x-ray measurements in terms of the temperature parameter Y of the Debye-Waller factor, the Debye characteristic temperature Θ_M and the mean square displacement of the atoms. The theoretical results are found to be in reasonably satisfactory agreement with the experimental values.

Introduction

It has been shown experimentally that the intensity of x-ray diffraction maxima decreases with rising temperature on account of thermal vibrations. An explanation of this effect was first given by Debye and subsequently modified by Waller who showed that this decrease could be expressed by an exponential factor $\exp\{-2W\}$, often referred to as the Debye-Waller factor. The exponent $2W$ is directly related to the mean-square displacement of the atoms $\overline{u^2}$ and correlates with many other solid state phenomena such as neutron scattering, electrical conductivity and melting. The exponent $2W$ has normally been calculated by the hypothetical Debye model applied to lattice vibrations of crystals and is expressed as a known function involving the characteristic temperature Θ. This method has been shown to be inadequate[1] and therefore necessitates a more realistic approach to compute the Debye-Waller factor of cubic structures.

However, in the recent past various phenomenological models have been applied by a number of workers (Chandra and Hemkar[2], Satya Pal[3], in explaining the lattice dynamics of both bcc and Gupta[7] to compute the exponent $2W$ using their vibration spectra. But these approaches min the symmetry requirements of the crystal and hence the frequencies obtained are not periodic in reciprocal space. In addition, in summing the series, the term corresponding to $k=0$ has been neglected by these workers because the value of the wave vector in this case corresponds to a non-vibrating lattice for which the Debye-Waller exponent vanishes. Therefore, the contribution of the central part of the Brillouin zone has not been taken into account in these calculations. Hence, not much reliance can be bestowed upon their computed $2W$ values.

Recently Shukla et al.[8] suggested a modification in the Clark, Gazis and Wallis (CGW) model[9] for fcc metals, in order to make it more in accord with the physical situation, by incorporating the effect of the conduction electrons. They have used this approach in studying the phonon dispersion of copper and have claimed its superiority over other phenomenological and angular force models as better results are obtained with it. Quite recently a similar improvement in the CGW angular force model for the bcc metals has also been made by the present authors[10, 11] and was used successfully for the lattice dynamical study of a number of bbc metals[10–13]. The marked success of this modified CGW scheme in explaining the lattice dynamics of both bcc and fcc metals has prompted us to follow the same procedure in studying the temperature dependence of the Debye-Waller factor exponents, the Debye characteristic temperature and the mean square displacements of the atoms in the five fcc metals, viz. Cu, Ag, Au, Al and Ni.

This approach takes into account the electron-ion interaction term of the Krebs model[14] along with the ion-ion central and ion-ion angular interaction as in the CGW model. The angular and central interactions are assumed to be effective only up to second neighbours. This model is superior to the above mentioned models in the respect that it satisfies the symmetry requirements of the lattice.

Theory

In the present calculations the contribution to the Debye-Waller exponent W is assumed to be consisting of two parts:

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(a) The phonon dependent part W' which is obtained from the knowledge of the whole vibration spectrum.

(b) The part W'' which is the contribution of the central part of the Brillouin zone, corresponding to $k=0$ and is evaluated in the Debye approximation.

(a) In the harmonic approximation, the exponent of the Debye-Waller factor W' is related to the mean square displacement of the atoms and is written as (James [15])

$$2\,W' = \langle |\mathbf{K}\cdot\boldsymbol{u}(n)|^2\rangle \tag{1}$$

where $\boldsymbol{u}(n)$ is the displacement of the nth atom and $\mathbf{K}$ denotes the difference of the initial and final wave-vectors of the wave. From the knowledge of the time dependence of the atomic displacements and the average energy of the phonon in mode $\boldsymbol{q}$, the amplitude $\boldsymbol{U_q}$ of the mode $\boldsymbol{q}$ can be written as

$$|\boldsymbol{U_q}|^2 = (n_{\boldsymbol{q}} + \tfrac{1}{2})\hbar/m\,N\cdot\omega_{\boldsymbol{q}}\,. \tag{2}$$

Here m is the mass of the atom, N the total number of unit cells in the crystal, $\hbar$ Planck's constant divided by 2π, $\omega_{\boldsymbol{q}}$ the frequency of the normal mode of wave-vector $\boldsymbol{q}$, and $n_{\boldsymbol{q}}$ the average occupation number of $\boldsymbol{q}$ lattice mode given by

$$n_{\boldsymbol{q}} = [\exp\{\hbar\,\omega_{\boldsymbol{q}}/k_{\mathrm{B}}\,T\} - 1]^{-1} \tag{3}$$

where T is the absolute temperature and k_{B} Boltzmann's constant. With the help of Eq. (1) and (2) we get

$$2\,W' = \frac{\hbar}{m\,N}\sum_{\boldsymbol{q},j}\frac{(\mathbf{K}\cdot\boldsymbol{e}_{\boldsymbol{q},j})^2\,(n_{\boldsymbol{q},j}+\frac{1}{2})}{\omega_{\boldsymbol{q},j}}\,, \tag{4}$$

where j is the polarization index and $\boldsymbol{e}_{\boldsymbol{q},j}$ the polarization vector of the $(\boldsymbol{q},j)$ lattice mode. The summation over $\boldsymbol{q}$ extends over all normal vibrations of the crystal. For a monatomic cubic crystal, the polarization factor $)\mathbf{K}\cdot\boldsymbol{e}_{\boldsymbol{q},j})^2$ may be replaced by its average value outside the summation and Eq. (4) can be rewritten as

$$2\,W' = \frac{8\,\lambda^2\,\hbar}{3\,m\,N}\left(\frac{\sin\Theta}{\lambda}\right)^2\sum_{\boldsymbol{q},j}\frac{1}{\omega_{\boldsymbol{q},j}}\,\mathrm{Coth}\left(\frac{\hbar\,\omega_{\boldsymbol{q},j}}{2\,k_{\mathrm{B}}\,T}\right). \tag{5}$$

Here Φ is the glancing angle of the incidence and λ the wave length of the incident wave.

(b) Since all the modes are acoustic for a monatomic lattice, ω is zero when $k=0$. Following Barron and Smith [16] we can avoid this singularity in the sum by replacing the contribution to the sum at $k=0$ with an integration of the energy term, $[E/4\,\lambda^2\,v^2$ (of the mode of lattice vibrations)$]$

$$E = \hbar\,\omega\,[(\exp\{\hbar\,\omega/k_{\mathrm{B}}\,T\} - 1)^{-1} + \tfrac{1}{2}] \tag{6}$$

over a sphere of volume equal to 20^{-3} of the Brillouin zone near $k=0$. It is assumed that the frequency is given in this region by the average velocity times the wave-vector. The zero phonon contribution W'' is then written as

$$W'' = \frac{a^3}{3\,m}\{D(C_{\mathrm{L}}) + 2\,D(C_{\mathrm{T}})\}\left(\frac{\sin\Theta}{\lambda}\right)^2 \tag{7}$$

where

$$D(C) = \frac{1}{C^2}\int_0^{2\lambda k_{\max}} 4\,\lambda^2\,\hbar\,c\,k\,\mathrm{d}k\left[\left(\exp\left(\frac{\hbar\,\omega}{k_{\mathrm{B}}\,T}\right) - 1\right)^{-1} + \frac{1}{2}\right]. \tag{8}$$

C_{L} is the average velocity for the longitudinal phonons and C_{T} is that for the transverse phonons in this region. $k_{\max}$ is the radius of the sphere of integration and is given by

$$\frac{4\,\lambda}{3}\,k_{\max}^3 = 20^{-3}\ \text{(volume of the Brillouin zone)}$$

or
$$k_{\max} = \frac{1}{20\,a}\left(\frac{3}{\lambda}\right)^{1/3}\ \text{(for fcc) metals}\,. \tag{9}$$

$D(C)$ can be reduced to

$$D(C) = P\left(\frac{1}{x}\int_0^x\frac{t\,\mathrm{d}t}{e^t - 1}\right) + \frac{1}{4}\,p\,x\,, \tag{10}$$

where $P = 4\,\lambda^2\,k_{\mathrm{B}}\,T'\,k_{\max}/C^2$, $x = 4\,\lambda^2\,\hbar\,k_{\max}\,C/k_{\mathrm{B}}\,T$

and $t = 2\,\lambda\,\hbar\,k\,C/k_{\mathrm{B}}\,T$.

The value of the integral contained in the expression for $D(C)$ can be substituted from standard tables. The Debye-Waller factor W is now calculated from the Eqs. (5) and (7). For a Debye model of the solid, the temperature dependence of the Debye-Waller exponent can be written as

$$2\,W = 48\,\lambda^2\,\hbar^2\,T/m\,k_{\mathrm{B}}\,\Theta_{\mathrm{M}}^2\,\{\Phi(x) + x/4\}\,(\sin\Theta/\lambda)^2\,. \tag{11}$$

Here Θ_{M} represents the effective x-ray characteristic temperature, $\Phi(x)$ the usual Debye integral function and $x = \Theta_{\mathrm{M}}/T$.

Similarly, in harmonic approximation, the mean square displacement of the atoms $\overline{u^2}$ is given by the following relation

$$\overline{u^2} = \frac{3}{8\,\lambda^2}\left(\frac{\lambda}{\sin\Theta}\right)^2 W\,. \tag{12}$$

Numerical Computation

The Debye-Waller exponent $2\,W$ for the five fcc metals has been calculated by Blackmann's root sampling technique [17]. For this purpose it is essen-

Table 1. Physical constants for the metals used in the calculations.

Metal	Lattice constant [Å]	Elastic constants 10^{11} dyne/cm^2			References
		C_{11}	C_{12}	C_{44}	
Copper	3.603	17.62	12.49	8.18	Svensson et al. [18] Kittel [19]
Silver	4.060	13.15	9.73	5.11	Kamitakahara and Brockhouse [20] Kittel [19]
Gold	4.056	20.16	16.97	4.54	Kittel [19]
Aluminium	4.030	11.43	6.19	3.16	Kittel [19] Yannell et al. [21]
Nickel	3.514	26.12	15.08	13.17	Kittel [19] Birgeneau et al. [22]

tial to determine the vibration frequencies at a suitable number of points in an irreducible section of the first Brillouin zone. In order to keep the frequencies within limits and yet to have a fairly large sample, we have considered a mesh of 8000 wave vectors evenly distributed in the first Brillouin zone. From the symmetry requirements the eight thousand points were reduced to 262 non-equivalent points, including the origin, lying within 1/48 of the irreducible part of the Brillouin zone.

The secular equation determining the lattice frequencies of the metals was solved for these non-equivalent points and thus the complete vibrational spectrum was obtained. In evaluating the summation in Eq. (5), we divided the whole vibrational spectrum into a number of frequency intervals and assigned appropriate weights to each interval. The mean square displacement of the atoms $\overline{u^2}$ and the Debye characteristic temperature Θ_M at different temperatures were evaluated from Eqs. (12) and (11), respectively.

The elastic constants and other relevant parameters used in the calculations are given in Table 1.

Comparison with Experiments

The comparison of the calculated and experimental Debye-Waller factors may be made in terms of a temperature parameter Y defined by

$$Y = \log_{10} e (\lambda/\sin \Theta)^2 \, (2\,W_{T_0} - 2\,W_T)$$

where W_{T_0} and W_T are the Debye-Waller exponents at temperature T_0 and T respectively. This quantity is independent of λ and Θ and is directly accessible from the measured x-ray intensities of the Bragg reflections. Now

$$I_T/I_{T_0} = \exp\{-2\,W_T\}/\exp\{-2\,W_{T_0}\}\,,$$

where I_T and I_{T_0} are the x-ray intensities at temperatures T and T_0 respectively. Hence, we have $Y = (\lambda/\sin\Theta)^2 \log_{10}(I_T/I_{T_0})$. The calculated values of Y, $\overline{u^2}$ and Θ_M at different temperatures for copper, silver, gold, aluminium and nickel are presented in Figs. 1 – 5, respectively. For comparison, the corresponding values deduced from various x-ray measurements have also been plotted.

Copper. The temperature dependence of the Debye-Waller factor of copper from x-ray intensity mea-

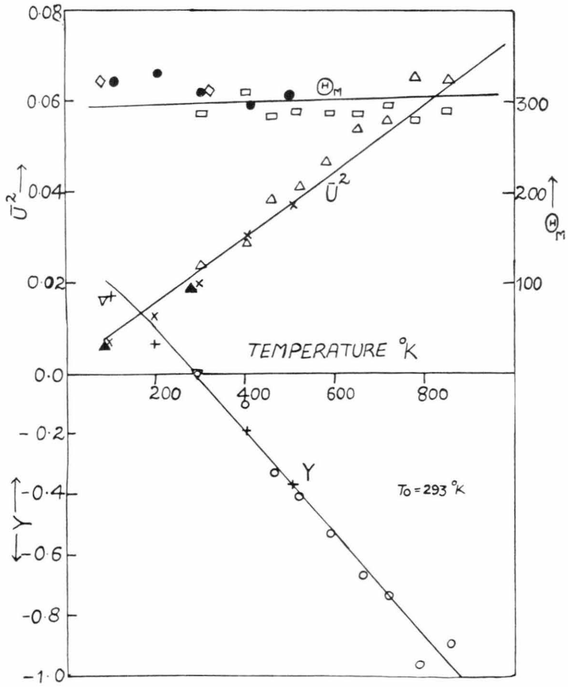

Fig. 1. Variation of Y, $\overline{u^2}$ and Θ_M with temperature for copper. Solid curves show the present calculations. Experimental points: + × ● Flinn et al.; ○ △ □ Owen and Williams; ▽ ▲ ◇ Chipman and Paskin.

surements has been investigated by Owen and Williams[23], Flinn et al.[24] and Chipman and Paskin[25]. Flinn et al. have measured the integrated intensities of the (600), (800) and (1000) reflections from a single copper crystal at selected temperatures in the temperature range 4.2 to 500 K. The observations have been expressed in terms of the effective Debye temperature Θ_M. For the sake of the present comparison, we have calculated the values of

$$(\lambda/\sin\Theta)^2 \log_{10}(I_T/I_{T_0})$$

as a function of temperature from the corresponding values of Θ_M. Measurements of Owen and William's covered the temperature range from 293 to 900 K for microphotometric measurements on the lines of the x-ray structure spectra of powder specimens. They have tabulated the values of Y at selected temperatures. Both these measurements are consistent and are in agreement with those of Chipman and Paskin[25] at room and liquid nitrogen temperatures on copper powder. The values of Y, $\overline{u^2}$ and Θ_M deduced from the measurements of all these workers are plotted in Figure 1. The theoretical results are in excellent agreement with experimental values.

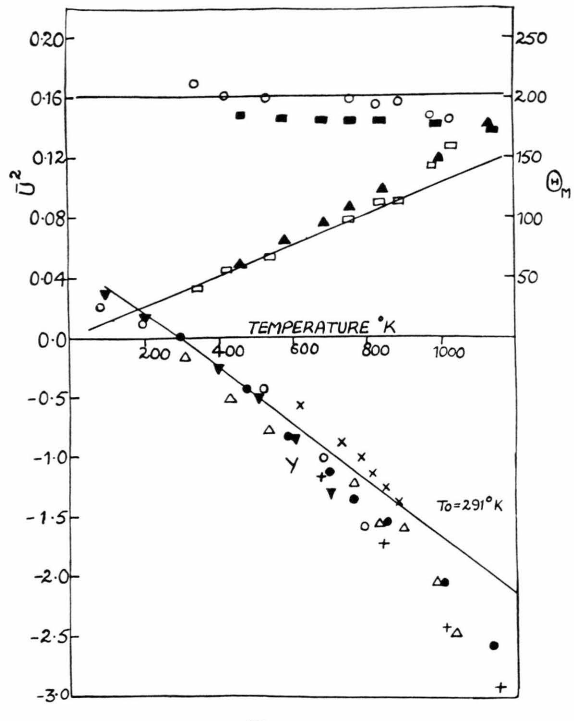

Fig. 2. Variation of Y, $\overline{u^2}$ and Θ_M with temperature for silver. Solid curves show the present calculations. Experimental points: ○ Boskovits et al.; × Andressen; + Spreadborough and Christian; ● ▲ ■ Haworth; ○ △ □ Simerska; ▼ Alexopoulos.

Silver. The effect of temperature on the intensity of x-ray reflections for silver have been investigated by different workers[26-31] by using slightly different techniques. Andriesson[27] used silver powder and carried out the intensity measurements over different diffraction lines in the temperature range 291 to 875 K. His results are, however, not sufficiently accurate as they were affected by the change in extinction effects and the oxidation of the surface of the scattering material. Boskovits et al.[26] observed the integrated intensities of diffraction lines (111) and (422) from a silver wire at six different temperatures between 81 and 774 K. Their values show a rapid decrease of Θ_M with increasing temperature beyond 290 K which is in discrepancy with other workers. Spreadborough and Christian[28] have measured the peak intensities of the diffracted powder lines (511), (333), (422) and (420) in the temperature range 273 to 1343 K. Their Θ_M values are about 22% lower than those obtained by Andriessen. Haworth[29] has measured the peak as well as integrated intensities in the temperature range 286 – 1100 K taking account of the thermal contribution to Bragg's peaks and repeatedly annealing the speciment at 1220 K. His results are in excellent agreement with the measurements of Simerska[30] carried out in the temperature range 291 – 1033 K but are in considerable disagreement with those of other workers. Alexopoulos et al.[31] have recently studied the temperature variation of intensity of x-ray reflections from a flat powder silver sample in the temperature range 100 – 700 K. The values of Y, $\overline{u^2}$ and Θ_M deduced from the measured intensities of all these workers have been considered for the present comparison with our theory in Figure 2. The agreement of the theoretical values of Y with experiment is satisfactory upto about 700 K, but above this temperature the theory disagrees with the experiment and the discrepancy increases with rise in temperature. The computed values of $\overline{u^2}$ and Θ_M are in reasonably satisfactory agreement with the experimental values of Haworth and Simerska.

Gold. Measurements of the temperature variation of integrated intensities of x-ray reflections from gold have been reported by Owen and Williams[23], Alexopoulos et al.[31] and Synecek et al.[32]. The observations of Owen and Williams have been taken

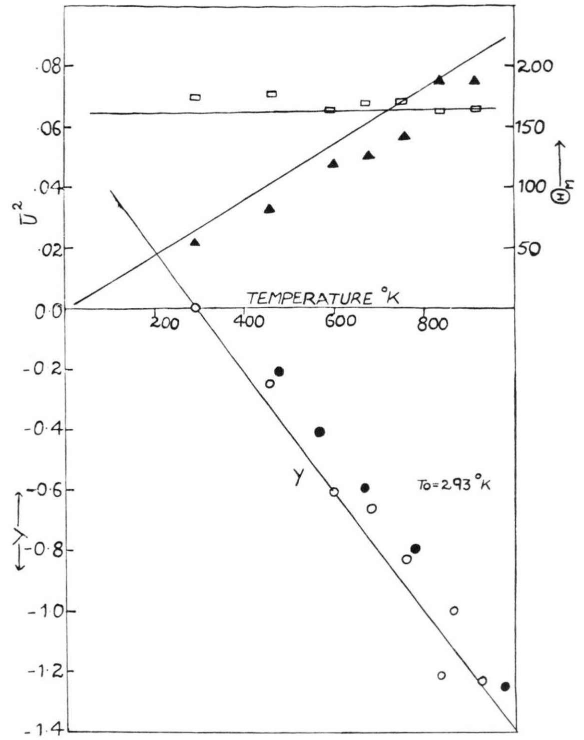

Fig. 3. Variation of Y, $\overline{u^2}$ and Θ_M with temperature for gold. Solid curves show the present calculations. Experimental points: ○ ▲ □ Owen and Williams; ● Synecek et al.

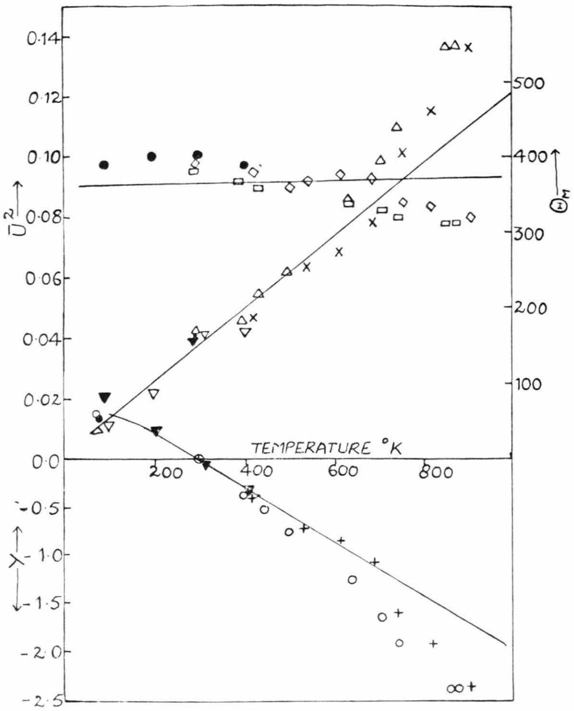

Fig. 4. Variation of Y, $\overline{u^2}$ and Θ_M with temperature for aluminium. Solid curves show the present calculations. Experimental points: × ▼ James et al.; + × ◇ Owen and Williams; ○ △ □ Chipman; ▼ ▽ ● Flinn and Mc Manus.

in the temperature range 293 to 921 K and those of Alexopoulos from 100 to 800 K. X-ray integrated intensities of gold single crystal from room temperature to 1150 K for the reflections (333), (335), (444), (553), (446), (555), (664), (557) and (666) have been measured by Synecek et al. who expressed their results in terms of $\overline{u^2}$ and Θ_M. The measurements of all these workers are plotted in Fig. 3 with $T_0 = 293$ K along with our calculated values. A satisfactory agreement between theory and experiment is observed throughout the temperature range studied.

Aluminium. X-ray intensity measurements for aluminium at different temperatures have been reported by Backhurst[33], Collins[34], James et al.[35], Owen and Williams[23], Chipman[36] and Flinn and McManus[37]. The observations of Backhurst and Collins are not accurate because of recrystallization and annealing effects due to heating and differ appreciably from other workers. The first reliable measurements of the integrated intensities of x-rays are those of James et al. at liquid air and room temperature. Owen and Williams have made observations in the temperature range 293 – 900 K and tabulated the Y values at selected temperatures. Chipman has made his measurements in the temperature range 60 – 880 K and corrected them for thermal diffuse scattering. Flinn and McManus measured the x-ray Debye Θ_M of aluminium in the temperature range 4.2 – 400 K using a flat single crystal with faces cut parallel to (100). For the present comparison, we have chosen the data of James et al., Owen and Williams, Chipman and Flinn and McManus with $T_0 = 293$ K and compared them with our theoretical values as plotted in Figure 4. The calculated Y values are found to be in good agreement with the experimental values upto 600 K and thereafter the discrepancy increases with rising temperature.

The calculated results for $\overline{u^2}$ and Θ_M show reasonably satisfactory agreement with experiments.

Nickel. X-ray measurements of the Debye-Waller factor for nickel have been made by Simerska[38] and Wilson et al.[39] Simerska carried out his ex-

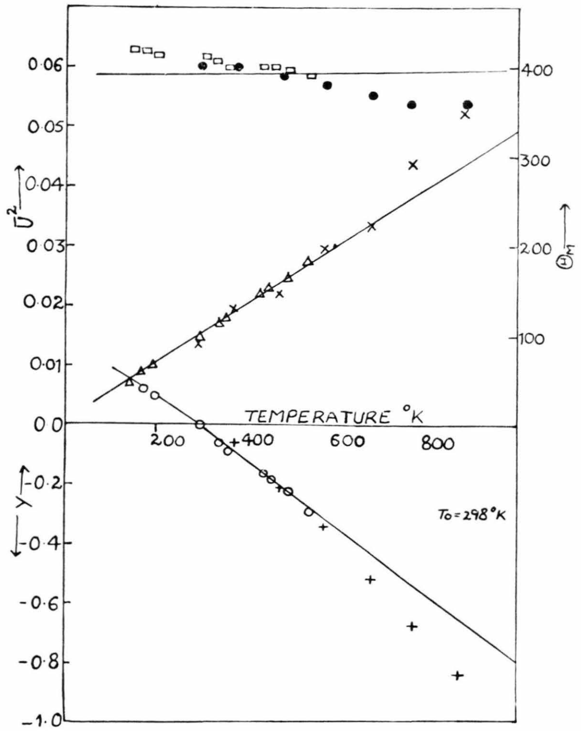

Fig. 5. Variation of Y, $\overline{u^2}$ and Θ_{M} with temperature for nickel. Solid curves show the present calculations. Experimental points: + × ● Simerska; ○ △ □ Wilson et al.

perimental work from room temperature to 873 K using a flat polycrystalline sample of nickel, Wilson et al., on the other hand, have measured the integrated intensities in the temperature range 100 to 500 K and corrected the data for thermal diffuse scattering and lattices parameter. These results agree with each other and have been plotted in Fig. 5 with $T_0 = 298$ K. The calculated values of Y, $\overline{u^2}$ and Θ_{M} agree satisfactorily with the experimental values.

Discussion

The present study shows that the Debye-Waller temperature parameter Y obtained from the modified CGW angular force model offers a satisfactory explanation for the observed temperature variation of the x-ray intensities of Bragg reflections up to a certain temperature. However, at higher temperatures the experimental values of $2W$ are consistently higher than the theoretical ones and this discrepancy increases with temperature. The disagreement between theory and experiment at high temperatures may be attributed to the neglect of the lattice expansion (Zener and Bilinsky [40]) and other anharmonic effects (Hahn and Ludwig [41], Cowley [42], Maradudin and Flinn [43], Slater [44] and Wolfe and Goodman [45]). Anharmonic contributions vary mostly as the square of the absolute temperature and become predominant at high temperatures. In the calculations no account has been taken of the temperature variation of the elastic constants and the lattice parameter. With increasing temperture the normal mode frequencies decrease because of thermal expansion. This effect depends upon the Grüneisen parameter which changes with lattice frequencies and temperature. According to studies made by various workers it has been seen that frequency distributions of fcc metals can be approximated by two peaks, the lower frequency peak being of larger area. At higher temperatures the Debye-Waller exponent depends the lattice frequencies as $\sum_{\boldsymbol{q},j} \omega_{\boldsymbol{q},j}^{-2}$. The lower frequency peak therefore contributes much more to $2W$ than the higher frequency peak and therefore largely governs the intensity of x-ray reflections. Since the Grüneisen parameter for this peak and its temperature variation is not known, no quantitative calculation of the thermal expansion effect on the x-ray intensities has been possible.

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