Debye-Waller factors of nickel and chromium

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The temperature variation of the Debye–Waller factors of nickel and chromium has been calculated from the vibrational spectrum derived from the lattice dynamical model of Krebs. The frequency spectra have been computed with the help of Blackman's root sampling technique for a discrete subdivision in the reciprocal space. The theoretical results are in reasonable agreement with the experimental data.

In the nonresonant elastic scattering of slow neutrons, x rays, etc., from scatterers which are bound in crystals, the fall in the intensity on account of lattice thermal motion is governed by an exponential factor e^{-2W}, called the Debye–Waller factor. The experimental data on the Debye–Waller factor have often been interpreted using simple Debye model for the vibration spectrum and results are expressed in terms of a known function involving the Debye characteristic temperature. The detailed theoretical and experimental information about the lattice dynamics of solids has proved the inadequacy of this approach and it is all the more imperative, nay pertinent to compute the Debye–Waller factor by using a more realistic phonon spectrum than the Debye one.

During the last few years, a number of lattice dynamical models have been proposed, in which the electron gas has been taken into account in the various approximate ways.[1] However, because of the lack of the translational invariance of the crystal lattice all these models violate the symmetry requirements of a cubic lattice.[2] Recently Krebs[3] has proposed an elastic force model which takes into account explicitly the effect of conduction electrons on the lattice vibrations and also satisfies the symmetry requirements of a cubic lattice. The Krebs model has successfully explained the lattice dynamical behavior of a number of cubic metals.[4-7] The results of the calculations of the temperature variation of the Debye–Waller factors of nickel (face-centered cubic) and chromium (body-centered cubic) on the basis of the lattice dynamical model of Krebs are presented herein.

In the quasiharmonic approximation, the exponent of the Debye–Waller factor which is related to the mean square displacement of the atoms,[8] can be written in terms of eigenvalues and eigenvectors of the phonons as

$$2W = \frac{\hbar}{mN} \sum_{q,p} \frac{(S \cdot e_{q,p})^2}{\omega_{q,p}} \left[\frac{1}{2} + \frac{1}{\exp(\hbar\omega_{q,p}/k_B T) - 1} \right], \qquad (1)$$

where m is the mass of an atom in the lattice, N is the total number of unit cells in the crystal, $\omega_{q,p}$ is the angular frequency of a phonon of wave vector q and polarization p, $e_{q,p}$ is the polarization vector of the $q-p$ lattice mode, k_B is the Boltzmann constant, and T is the absolute temperature. For a lattice with cubic symmetry,[9] the polarization factor $(S \cdot e_{q,p})^2$ can be replaced by its average value outside the summation. If $G(\omega)$ is the frequency distribution function for the phonon giving the number of the vibrational modes in the frequency interval ω and $\omega + d\omega$, Eq. (1) can be written as

$$2W = \frac{8\pi^2 \hbar}{3mN} \left(\frac{\sin\theta}{\lambda}\right)^2 \int_0^{\omega_{max}} \frac{G(\omega)}{\omega} \coth\frac{\hbar\omega}{2k_B T} \, d\omega, \qquad (2)$$

where θ is the glancing angle of incidence, λ is the wavelength of the incident radiation, and ω_{max} is the maximum frequency of the vibrational spectrum.

The calculation of the temperature variation of the Debye–Waller exponent $2W$ from Eq. (2) has been made by Blackman's root-sampling technique.[10] For this purpose, in order to get a fairly large survey of frequencies, the reciprocal space has been divided into miniature cells with axes one-twentieth of the length of the reciprocal lattice cell, giving thus a mesh with the symmetry of the reciprocal lattice of 8000 wave vectors evenly distributed in the first Brillouin zone. From the symmetry considerations, the vibration frequencies are calculated from the roots of the Krebs[3] secular determinant for the nonequivalent points lying within the 1/48th irreducible portion of the first Brillouin zone. Including the origin, there are 256 such nonequivalent points for a body-centered lattice and 262 points for the face-centered cubic structure which by symmetry operations suffice to generate the 8000 points in the first Brillouin zone. Each frequency is weighted according to the symmetrically equivalent points. The vibration spectrum is divided into frequency intervals of 0.1×10^{13} rad/sec and the midpoint of each frequency step is taken

TABLE I.

	Elastic constants (10^{12} dyn cm^{-2})			Lattice constant (Å)	Ionic mass (a.m.u.)
	C_{11}	C_{12}	C_{44}		
Nickel[a]	2.460	1.500	1.220	3.5239	58.710
Chromium[b]	3.500	0.678	1.008	2.8845	51.996

[a]Reference 11.   [b]Reference 12.
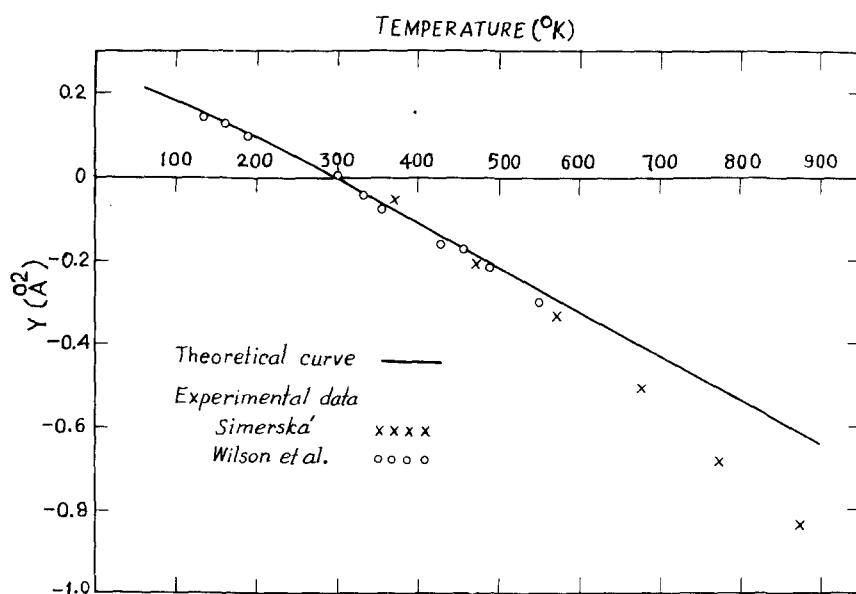
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TEMPERATURE (°K)

FIG. 1. The temperature variation of the Debye–Waller factor parameter of nickel.

as the representative of that interval with statistical weights equal to the number of frequencies lying within it. In the calculation, the value of the screening parameter for the electron–ion interaction occurring in the Krebs model has been taken to be 0.584. It has been found that good agreement with the experimental data is obtained if the screening constant is treated as an adjustable parameter, changing gradually from the Bohm–Pines to that of the Thomas–Fermi value as one goes from the lighter to heavier atoms. Using the frequency spectrum the Debye–Waller factor exponent $2W$ has been calculated by the numerical integration of Eq. (2). The numerical values of the elastic constants and other relevant data used in the calculation are listed in Table I.

The results have been compared with the experimental data in terms of the Debye–Waller temperature parameter Y which is independent of λ and θ–being completely determined by the vibrational spectrum and is easily accessible from the x-ray intensities of the Bragg re-

flections. If I_T and I_0 are the measured integrated intensities of a given reflection from the crystal at temperatures T and T_0, respectively, then

$$I_T/I_0 = \exp(-2W_T)/\exp(-2W_0),$$

which gives

$$\frac{\lambda^2}{\sin^2\theta}\ \log_{10}\frac{I_T}{I_0} = 2\ \log_{10}e\ \frac{\lambda^2}{\sin^2\theta}\ (W_0 - W_T).$$

$$= Y \tag{3}$$

The numerical values of the right hand side in the above equation are calculated with the help of Eq. (2). The results of the theoretical calculations of the temperature variation of the Debye–Waller factor for nickel together with the experimental data of Simerská[13] and Wilson et al.[14] have been plotted in Fig. 1. It is seen that the calculations are in reasonable agreement with the experimental values. However, the discrepancy between the theory and experiment increases with the rise
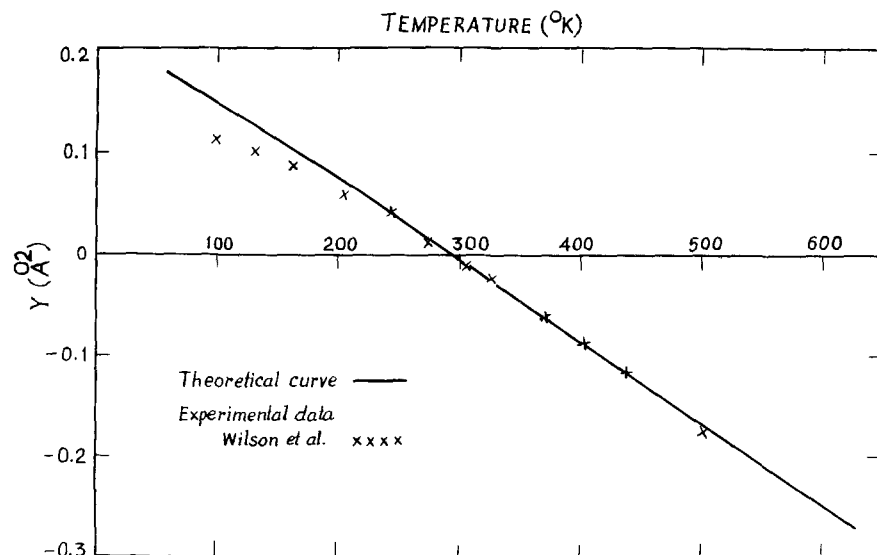
TEMPERATURE (°K)

FIG. 2. The temperature dependence of the Debye-Waller factor parameter of chromium.

in temperature. The theoretical results of the temperature dependence of the Debye–Waller factor of chromium have been compared with the experimental results of Wilson et al.[14] in Fig. 2. The theoretical values compare well with the experimental data throughout the temperature range studied.

The present investigation shows that the Krebs model gives a reasonable description of the temperature variation of the Debye–Waller factors of nickel and chromium. The agreement between the theory and experiment for chromium is surprisingly good. The discrepancy between the theoretical results and the experimental data can be attributed to the neglect of the temperature variation of the vibrational frequencies[15] and to the other anharmonic effects.[16-20] Moreover, the temperature variation of the elastic constants and the lattice parameter have not been taken into account in the present study. With the rise in temperature, lattice frequencies are diminished because of the lattice expansion. This effect depends upon the Grüneisen parameter which varies with temperature. Some attempts[21-23] have been made to estimate this effect in terms of the Grüneisen parameter but a detailed discussion had not been possible. A detailed study incorporating the Grüneisen parameter and the anharmonicity in lattice vibrations though very much desired for the interpretation of the Debye–Waller factor of solids, has not yet been attempted.

ACKNOWLEDGMENTS

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