

Reprinted from THE PHYSICAL REVIEW, Vol. 131, No. 2, 528-529, 15 July 1963  
Printed in U. S. A.

## Temperature Dependence of the Debye-Waller Factor for Copper and Aluminum

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(Received 11 March 1963)

The Debye-Waller factor for copper has recently been determined by Flinn *et al.* by making x-ray intensity measurements from 4–500°K. Flinn *et al.* were able to explain their results using a central force model for the copper lattice. However, it is well known that this model is inadequate in explaining the vibrational properties of the copper lattice over the entire wavelength region. Since various sets of force constants have been proposed for copper, we calculated the Debye-Waller factor for these sets. It is found that with the present experimental accuracy it is impossible to select between them. Calculations are also given for aluminum using Walker's atomic force constants.

### INTRODUCTION

RECENTLY, Flinn *et al.*<sup>1</sup> reported on the x-ray determination of the Debye-Waller factor for copper. They were able to interpret the experimental results in terms of a central force model (C.F.) including first- and second-neighbor interactions. Since the frequency spectrum is known for copper and was analyzed first by Jacobsen<sup>2</sup> using a full tensor model and second by Lehman *et al.*<sup>3</sup> using a special case of the full tensor model, it is interesting to compare the Debye-Waller factor predicted from these various sets of force constants. We have also calculated the Debye-Waller factor using the tensor force constants computed by White<sup>4</sup> from what amounts to a first-principles calculation using Feynman's theorem. Calculations of the Debye-Waller factor for aluminum are made using Walker's<sup>5</sup> tensor force constants which were determined from experimental dispersion curves. The method of calculation with the results is presented in Sec. II.

### RESULTS

The Debye-Waller factor for a cubic crystal can be written in the following manner:

$$f = e^{-2W}, \quad (1)$$

TABLE I. Atomic force constants for copper (in units of  $10^4$  dyn  $\text{cm}^{-1}$ ).

Force constant	Jacobsen	White	A-S	C.F.
$\alpha_1$	0.87	1.71	1.12	1.477
$\beta_1$	0.48	-0.24	-0.001	0
$\gamma_1$	1.25	1.66	1.12	1.477
$\alpha_2$	0.35	-0.13	-0.0227	-0.2753
$\beta_2$	-0.072	-0.07	-0.0105	0
$\alpha_3$	0.09	-0.01	0.1122	0
$\beta_3$	-0.022	+0.005	0.00345	0
$\gamma_3$	-0.015	+0.02	0.03625	0
$\delta_3$	0.06	+0.01	0.0725	0

<sup>1</sup> P. A. Flinn, G. M. McManus, and J. A. Rayne, Phys. Rev. 123, 809 (1961).

<sup>2</sup> E. H. Jacobsen, Phys. Rev. 97, 654 (1955).

<sup>3</sup> G. W. Lehman, T. Wolfram, and R. E. DeWames, Phys. Rev. 128, 1593 (1962).

<sup>4</sup> H. C. White, Phys. Rev. 112, 1092 (1958).

<sup>5</sup> C. B. Walker, Phys. Rev. 103, 547 (1956).

where

$$2W = RC(T), \quad (2)$$

and

$$C(T) = \frac{2}{3N} \sum_{\phi} (\bar{n}_{\phi} + \frac{1}{2}) \frac{1}{\hbar\omega_{\phi}}, \quad (3)$$

$\phi$  stands for two indices ( $q, j$ ) where  $q$  is the propagation vector and  $j$  the vibrational branch.  $\bar{n}_{\phi}$  is the average number of phonons in a given mode  $\phi$ .  $N$  refers to the total number of unit cells.  $R$  is the recoil energy of a free emitting atom.

TABLE II. Atomic force constants for aluminum (in units of  $10^4$  dyn  $\text{cm}^{-1}$ ).

Force constant	Walker	A-S
$\alpha_1$	0.845	0.845
$\beta_1$	-0.093	-0.09
$\gamma_1$	1.067	0.935
$\alpha_2$	0.214	0.2
$\beta_2$	0.04	-0.1
$\alpha_3$	0.027	0
$\beta_3$	-0.031	0
$\gamma_3$	0.01	0
$\delta_3$	-0.019	0

In order to calculate the constant  $C(T)$  the vibrational frequencies for an arbitrary propagation vector and branch are determined using a full tensor force model including third neighbors. The A-S (axially symmetric) calculation is done as a special case of the full tensor model.

Equation (3) is then evaluated by integrating over  $1/48$  of the Brillouin zone appropriate for the fcc structure. This portion of the Brillouin zone was divided into two regions. A triple Gaussian quadrature was used to evaluate the resulting integrals. The  $3 \times 3$  dynamical matrix was diagonalized by  $2 \times 2$  Jacobi rotation procedure at 1024 points in each region. Several checks were made to insure that the integration was independent of the order of the Gaussian quadrature. Table I gives the atomic force constants presently known for Cu which were used in the calculation of the Debye-Waller factor. Table II gives the atomic force constants

TABLE III.  $C(T)$  for copper (in units of  $10^2 \text{ eV}^{-1}$ ).

$T(^{\circ}\text{K})$	Flinn <i>et al.</i>		C.F. model	Jacobsen	White	A-S	Debye model $\theta_D=335^{\circ}\text{K}$
	Experimental values $\theta_D$	$C(T)$					
4	$320 \pm 10$	$0.544 \pm 0.02$	0.552	0.579	0.537	0.570	0.520
20	$320 \pm 10$	$0.588 \pm 0.02$	0.566	0.593	0.548	0.582	0.532
80	$320 \pm 10$	$0.755 \pm 0.04$	0.762	0.808	0.733	0.779	0.697
300	$315 \pm 10$	$2.17 \pm 0.14$	2.10	2.29	2.03	2.18	1.93
400	$300 \pm 10$	$3.14 \pm 0.25$	2.77	3.02	2.67	2.87	2.50

for aluminum determined by Walker from his experimental dispersion curves. The experimental and calculated temperature dependence of  $C(T)$  for copper is given in Table III. For comparison we calculated  $C(T)$

TABLE IV.  $C(T)$  for aluminum (in units of  $10^2 \text{ eV}^{-1}$ ).

$T(^{\circ}\text{K})$	Walker	Debye model ( $\theta_D=382^{\circ}\text{K}$ )
4	0.471	0.459
20	0.478	0.464
80	0.598	0.583
300	1.54	1.50
400	2.02	1.96

using a Debye model, the results of these calculations are also shown in this table.

From the results in Table III we conclude that the experimental determination of the Debye-Waller factor for pure host lattices of cubic symmetry is not sensitive enough to distinguish between the various models. Models inconsistent with experimental dispersion curves such as the C.F. model and White's full tensor model give essentially the same Debye-Waller factor.

The results of the calculations for aluminum using Walker's force constants are given in Table IV. Using the A-S force constants the values of  $C(T)$  are higher than Walker's by 1% at  $0^{\circ}\text{K}$  and by 4% at  $400^{\circ}\text{N}$ . Only a small difference is expected since Walker's constants are essentially axially symmetric.