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

R. E. DeWames, T. Wolfram, and G. W. Lehman
North American Aviation Science Center, Canoga Park, California

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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.*[1] 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[2] using a full tensor model and second by Lehman *et al.*[3] 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[4] 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[5] 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:

$$f=e^{-2W}, \tag{1}$$

where

$$2W=RC(T), \tag{2}$$

and

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

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

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

Force constant	Jacobsen	White	A–S	C.F.
α_1	0.87	1.71	1.12	1.477
β_1	0.48	−0.24	−0.001	0
γ_1	1.25	1.66	1.12	1.477
α_2	0.35	−0.13	−0.0227	−0.2753
β_2	−0.072	−0.07	−0.0105	0
α_3	0.09	−0.01	0.1122	0
β_3	−0.022	+0.005	0.00345	0
γ_3	−0.015	+0.02	0.03625	0
δ_3	0.06	+0.01	0.0725	0

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

Force constant	Walker	A–S
α_1	0.845	0.845
β_1	−0.093	−0.09
γ_1	1.067	0.935
α_2	0.214	0.2
β_2	0.04	−0.1
α_3	0.027	0
β_3	−0.031	0
γ_3	0.01	0
δ_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 3×3 dynamical matrix was diagonalized by 2×2 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

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[2] E. H. Jacobsen, Phys. Rev. 97, 654 (1955).
[3] G. W. Lehman, T. Wolfram, and R. E. DeWames, Phys. Rev. 128, 1593 (1962).
[4] H. C. White, Phys. Rev. 112, 1092 (1958).
[5] C. B. Walker, Phys. Rev. 103, 547 (1956).

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

T(°K)	Flinn *et al.* Experimental values θ_D	Flinn *et al.* Experimental values $C(T)$	C.F. model	Jacobsen	White	A-S	Debye model θ_D=335°K
4	320±10	0.544∓0.02	0.552	0.579	0.537	0.570	0.520
20	320±10	0.588∓0.02	0.566	0.593	0.548	0.582	0.532
80	320±10	0.755∓0.04	0.762	0.808	0.733	0.779	0.697
300	315±10	2.17 ∓0.14	2.10	2.29	2.03	2.18	1.93
400	300±10	3.14 ∓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 $C(T)$ 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°K and by 4% at 400°N. Only a small difference is expected since Walker's constants are essentially axially symmetric.

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

T(°K)	Walker	Debye model (θ_D=382°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

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Temperature Dependence and Anisotropy in the Debye-Waller Factor for White Tin

R. E. DEWAMES, T. WOLFRAM, AND G. W. LEHMAN
North American Aviation Science Center, Canoga Park, California
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The Debye-Walker factor, e^{-2W}, for tin is calculated using the A-S (axially symmetric) lattice dynamics model described in an earlier paper. The Debye continuum approximation is found to be unsatisfactory because the optical modes contribute significantly even at low temperatures. Calculated and experimental values determined from Mössbauer measurements are in excellent agreement in the temperature range from 0 to 300°K. Discrepancies above 300°K are attributed to higher order corrections such as anharmonicities and diffusion effects. In tin, the Debye-Waller factor depends upon the direction of gamma ray emission with the ratio $2W_x/2W_z$ varying from 1.1 to 1.2 for T=0°K and T=300°K, respectively. The calculated anisotropy in $2W$ is compared with available experimental data. Dispersion curves and values of $2W$ calculated using Rayne and Chandrasekhar elastic data are compared with those calculated using Mason and Bömmel elastic data. The effect of the relative motion of the two sublattices on the elastic properties of tin is discussed and found to be important for the elastic constants of Rayne and Chandrasekhar.

I. INTRODUCTION

THE probability of a gamma-ray emission without energy transfer to or from the lattice[1,2] and the temperature dependence of the atomic structure factor in the reflection of x rays[3] is given by

$$f=e^{-2W} \tag{1}$$

where $2W$ is related to the mean square displacement of an atom along a definite direction.

Since the experimental determination of f for tin has only been investigated through a study of the temperature dependence of recoil-less γ emission the constant $2W$ is defined for this specific case. Hence,

$$2W=R\sum_q\sum_j[\varrho^\alpha\cdot\mathbf{e}^\alpha(q,j)]^2 g[\omega(q,j)], \tag{2}$$

[1] R. L. Mössbauer, Z. Physik 151, 124 (1958).
[2] W. E. Lamb, Jr., Phys. Rev. 55, 190 (1939).
[3] R. W. James, *The Optical Principles of the Diffraction of X-Rays* (G. Bell and Sons, London, 1953).