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X-ray Debye Temperatures for Al, Nb and Pb

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Data from neutron scattering experiments are used to calculate X-ray Debye temperatures for Al, Nb and Pb.

In the literature there are many attempts to calculate Debye–Waller factors for metals from a more realistic phonon spectrum than a Debye model. In this note we present calculations of the X-ray Debye temperature Θ_M for the elements Al, Nb and Pb. We adopt standard theory and define Θ_M by (*e.g.* James, 1954)

$$\Theta_M^2 = \frac{3hT}{k_B}\Psi(\Theta_M/T) \times \frac{\int F(\nu)d\nu}{\int \frac{F(\nu)}{\nu}\left[\frac{1}{2}+\frac{1}{\exp(h\nu/k_BT)-1}\right]}. \quad (1)$$

$F(\nu)$ is the frequency distribution for the phonons and $\Psi(x)=\Phi(x)+x/4$ where $\Phi(x)$ is the usual Debye function. The Debye–Waller factor is then given by

$$M = \frac{6h^2T}{mk_B\Theta_m^2}\Psi(\Theta_M/T)\frac{\sin^2\theta}{\lambda^2}. \quad (2)$$

When $F(\nu)$ is known, equation (1) is easily solved by an iteration procedure. There are several calculations of Θ_M for aluminum. Some of them (Joshi, 1961; Kagan & Umanskii, 1962; Flinn & McManus, 1963; DeWames, Wolfram & Lehman, 1963; Mahesh, 1964) assume a model to get the phonon frequencies $\nu(\mathbf{q})$. Another method is to use a Born–von Kármán model to fit neutron data in the principal directions and then solve numerically for $\nu(\mathbf{q})$ in the rest of the Brillouin zone (Nicklow & Young, 1966; Gilat & Nicklow, 1966). Stedman and co-workers (Stedman & Nilsson, 1966; Stedman, Almqvist, Nilsson & Raunio, 1967), however, also measured $\nu(\mathbf{q})$ at off-symmetry points and so obtained a very accurate density of states $F(\nu)$. For Al this makes very little difference as compared with Born–von Kármán methods, but for lead there is a significant deviation (Stedman & Almqvist & Nilsson 1967). In Fig. 1 we give Θ_M obtained from equation (1) using Stedman's data. The frequency spectrum was measured at 80°K. It is therefore necessary to correct for the temperature dependence of the frequencies. Several attempts have been made to account for this shift (see Paskin (1957) and references therein). We assume that the lattice vibrations can be treated as harmonic oscillators also at higher temperatures. Stedman & Nilsson (1966) and Stedman, Almqvist, Nilsson & Raunio (1967) also measured $\nu(\mathbf{q})$ at 300°K. We use their results to find an average shift $\Delta\nu(\mathbf{q})/\nu(\mathbf{q})$ to be used in equation (1).

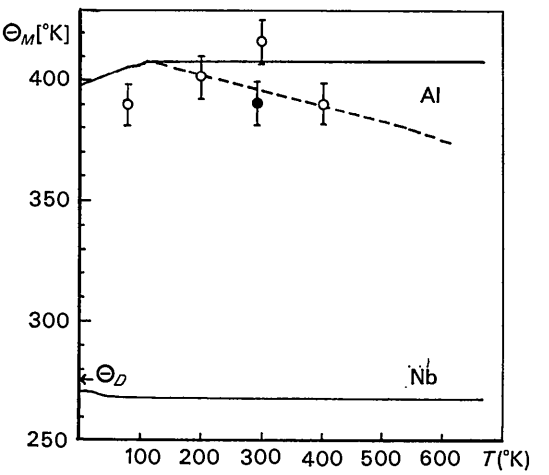

Fig. 1. Full-drawn curve: Θ_M for Al and Nb with constant $\nu(\mathbf{q})$. Dashed line: estimated correction from shift in $\nu(\mathbf{q})$ at higher temperatures. Experimental points with open circles from Flinn & McManus (1963) and with filled circle from Chipman (1960). Θ_D for Nb from van der Hoeven & Keesom (1964).

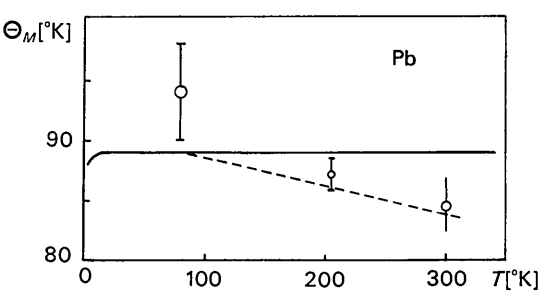

Fig. 2. Full-drawn line: Θ_M for Pb with constant $\nu(\mathbf{q})$. Dashed line: estimated temperature correction from shift in $\nu(\mathbf{q})$. Experimental points from Chipman (1960).

The average relative shift for Al at 300°K was estimated to 0·03. Assuming that the shift increases a little more than linear in T we have taken $\Delta\nu(\mathbf{q})/\nu(\mathbf{q}) = 0{\cdot}08$ at 600°K. This procedure is somewhat arbitrary but should nevertheless be as accurate as standard methods (Paskin, 1957) using Grüneisen's γ to obtain a linear shift in $\Theta(T)$. The full-drawn line in Fig. 1 shows Θ_M calculated with $\nu(\mathbf{q})$ as measured at 80°K, and the dashed line is with our temperature correction included. At low temperatures there will be some structure (*i.e.* Θ_M is not constant) due to the difference between a Debye model and the true phonon spectrum. We find that the size of the structure in Θ_M at low temperature is smaller than that obtained by Kagan & Umanskii (1962), and the high temperature limit of Θ_M is about 15°K higher than that of Flinn & McManus (1963) (without temperature correction). The agreement is excellent with the calculations of Nicklow & Young (1966) based on Stedman & Nilsson's (1966) measurements in principal directions and using a force-constant fitting (their $\Theta_{M'}$ is defined slightly different but equals Θ_M at high temperatures). Gilat & Nicklow (1966) also give $\Theta_{M'}$ from phonon measurements at 300°K, *i.e.* they take into account that the relative change in $\nu(\mathbf{q})$ with temperature is not the same for all phonons. Our estimated average $\Delta\nu(\mathbf{q})/\nu(\mathbf{q})$ leads to a good agreement with their results. In Fig. 1 we also give experimental values from measurements by Chipman (1960) and Flinn & McManus (1963).

The treatment for lead is also based on measurements by Stedman, Almqvist, Nilsson & Raunio (1967) and is completely analogous to that of aluminum. The estimated average shift $\Delta\nu(\mathbf{q})/\nu(\mathbf{q})$ at $T=$ 300°K is 0·06. Theoretical curves and experimental values from Chipman (1960) are shown in Fig. 2.

The density of states for niobium is taken from Nakagawa & Woods (1963) and is based on a force-constant fitting (eight neighbours) to neutron data. We have found no experimental data for Θ_M but mark $\Theta_D(T=0)$ as determined from heat capacity measurements.

In experiments Θ_M is inferred indirectly from intensity measurements in X-ray scattering. This makes the experimental Θ values very uncertain at low temperatures. Also the error bars are a somewhat arbitrary estimation. For a full discussion of this point see Chipman (1960). Our calculations are based on very accurate phonon data. The only (but important) uncertainty is the temperature dependence of the phonon spectrum. The results can make a good reference in later experimental studies. However the experimental data available do not allow for a more detailed comparison. The apparent discrepancy between theory and experiment (Figs. 1 and 2) is probably of no significance, as the error bars are only estimates.

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