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Phonon Dispersion Relations for Chromium and Tantalum

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Abstract

Phonon dispersion curves for the normal modes of vibration in chromium and tantalum are calculated along the symmetry directions [100], [110] and [111] using the five force-constant model of Behari and Tripathi (1970*a*). The results are compared with experimental values obtained from inelastic neutron spectroscopy and reasonably good agreement is found.

Introduction

Behari and Tripathi (1970*a*, 1970*b*, 1971*a*, 1971*b*) have investigated the lattice dynamics of several b.c.c. and f.c.c. metals using a modified form of a noncentral force model which takes into account the de Launay (1956) type angular force and the Sharma and Joshi (1963) type volume forces. Behari and Tripathi emphasized that the Cauchy discrepancy in metals arises from the angular interaction between a pair of ions and from the presence of the electron gas. Phonon angular frequencies calculated on this approach show good agreement with the recent experimental neutron-scattering data in almost all cases. The Behari–Tripathi model was also found to give a plausible account of the phonon dispersion relations and heat capacities in the transition metals α-iron, molybdenum and tungsten (Behari and Tripathi 1972) and of the temperature dependence of the Debye–Waller factors for the noble metals copper, silver and gold (Chandra and Hemkar 1973). We therefore consider it worth while to apply this model to other complicated cases of transition metals from the fifth and sixth columns of the Periodic Table.

In the present paper, we use the Behari–Tripathi model to calculate phonon dispersion relations in the transition metals chromium and tantalum along the symmetry directions [100], [110] and [111]. The motivation for this study was the recent appearance of detailed phonon dispersion curves for chromium (Møller and Mackintosh 1965; Shaw and Muhlestein 1971) and tantalum (Woods 1964) obtained from experiments on the coherent inelastic scattering of neutrons. The present study provides an additional check on the accuracy of the Behari–Tripathi model.

Secular Determinant

The secular equation determining the angular frequencies ω of the normal modes of vibration in a cubic metal can be written as

$$|M - m\omega^2 I| = 0, \tag{1}$$

where m is the mass of the atom and I is the unit matrix of order three. The elements

of the dynamical matrix M for a b.c.c. lattice are (Behari and Tripathi 1970*a*)

$$M_{11} = \tfrac{8}{3}(\alpha_1+2\alpha')(1-C_1C_2C_3)+4\alpha_2 S_1^2 + 2\alpha''\{2-\cos(2\pi k_2 a)-\cos(2\pi k_3 a)\}+2a^3\pi^2 k_1^2 G^2(qr_0)K_e, \quad (2a)$$

$$M_{12} = \tfrac{8}{3}(\alpha_1-\alpha')S_1S_2C_3+2a^3\pi^2 k_1 k_2 G^2(qr_0)K_e, \quad (2b)$$

where

$$C_i = \cos\pi a k_i, \quad S_i = \sin\pi a k_i, \quad \boldsymbol{q} = 2\pi\boldsymbol{k}, \quad G(x) = 3x^{-3}(\sin x - x\cos x). \quad (3)$$

The other quantities appearing in the expressions for M_{11} and M_{12} are: the radius r_0 of the Wigner–Seitz sphere, the lattice constant a, the phonon wave vector $\boldsymbol{k}$ (with $|\boldsymbol{k}| = \lambda^{-1}$), the radial force constants α_1 and α_2, the angular force constants α' and α'', and the force constant K_e corresponding to the bulk modulus of the electron gas. The latter quantities comprise the five force constants of the Behari–Tripathi model.

Table 1. Physical constants and calculated force constants for chromium and tantalum at room temperature

Physical constant	Chromium	Tantalum	Calculated force constant	Chromium	Tantalum
C_{11} (10^{10} Pa*)	35·000	26·100	α_1 (10^2 Pa*)	25·400	49·125
C_{12} (10^{10} Pa)	6·780	15·740	α_2 (10^2 Pa)	25·462	15·275
C_{44} (10^{10} Pa)	10·080	8·180	α' (10^2 Pa)	6·865	3·525
a (10^{-10} m)	2·880	3·303	α'' (10^2 Pa)	1·462	−5·212
ν_{T_1} (10^{12} Hz)	7·830	5·030	aK_e (10^2 Pa)	23·808	18·060
ν_{T_2} (10^{12} Hz)	8·000	3·780			

* Note that 1 Pa (pascal) ≡ 10 dyn cm^{-2}.

In the long-wavelength limit of equation (1), the radial and angular force constants may be evaluated by relating them to two elastic constants ($C_{11}-C_{12}$) and C_{44} and two zone-boundary frequencies (ν_{T_1} and ν_{T_2} both transverse) in the [100] and [111] directions respectively. The resulting expressions are:

$$a(C_{11}-C_{12}) = 2\alpha_2+4\alpha'+2\alpha'', \quad (4a)$$

$$aC_{44} = \tfrac{2}{3}\alpha_1+\tfrac{4}{3}\alpha'+2\alpha'', \quad (4b)$$

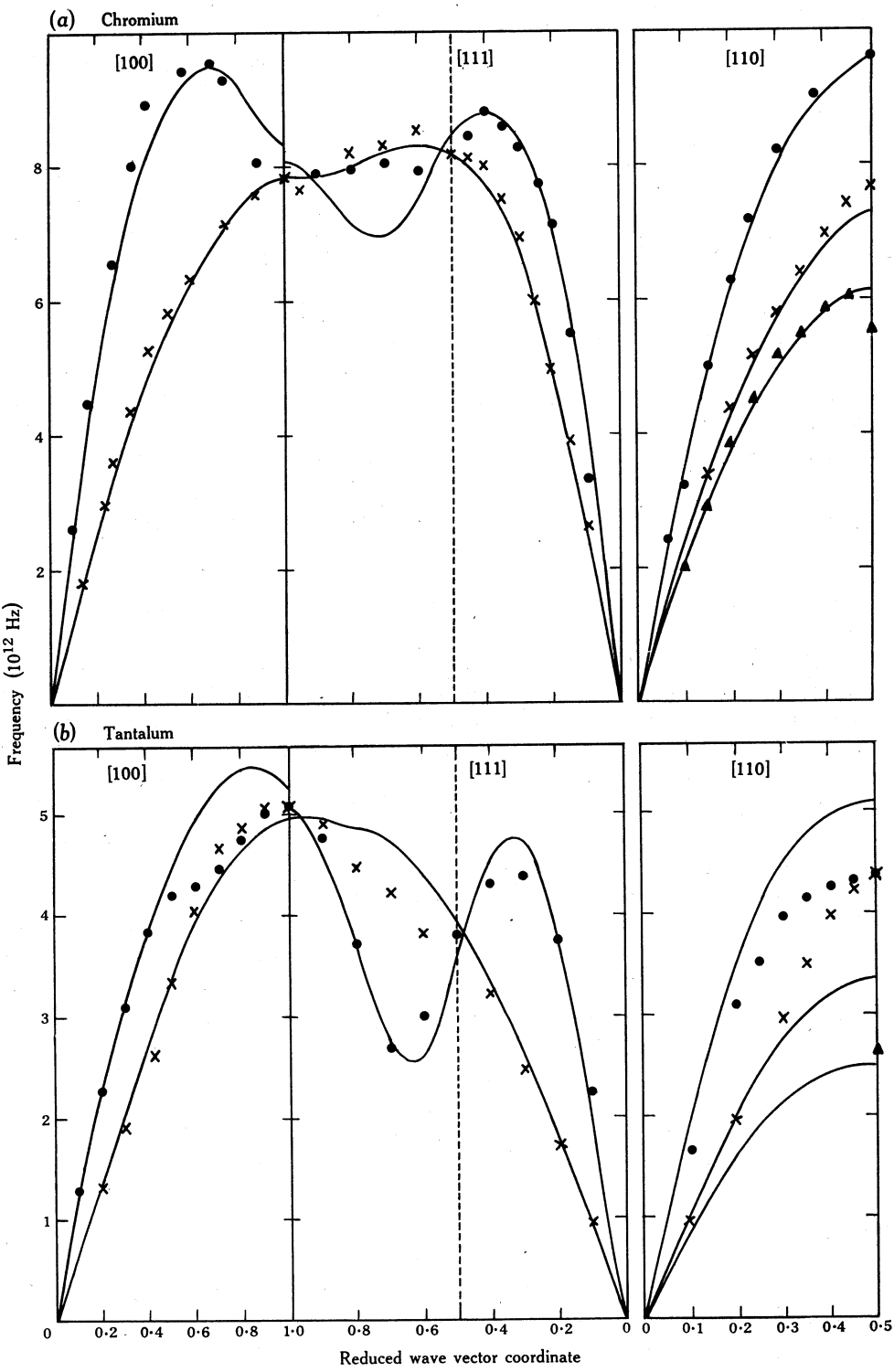
$$m\omega_{T_1}^2 = \tfrac{16}{3}(\alpha_1+2\alpha'), \quad (4c)$$

$$m\omega_{T_2}^2 = \tfrac{8}{3}(\alpha_1+2\alpha')+4\alpha_2+8\alpha''. \quad (4d)$$

The remaining force constant K_e may be determined from the equality

$$a(C_{12}-C_{44}) = -4\alpha'-4\alpha''+aK_e.$$

Fig. 1. Room temperature phonon dispersion curves along the indicated three symmetry directions in: (*a*) chromium, with experimental values by Shaw and Muhlestein (1971), and (*b*) tantalum, with experimental values by Woods (1964).


(a) Chromium
[100]
[111]
[110]
(b) Tantalum
[100]
[111]
[110]
Frequency (10^{12} Hz)
Reduced wave vector coordinate

The solutions of these equations determine the five force constants, which may then be substituted into equation (1) to yield the roots (frequencies) of the secular determinant for selected values of the phonon wave vector. Experimental values for the relevant elastic constants, zone-boundary frequencies and the lattice constant, together with the calculated values of the force constants, are listed in Table 1.

Results and Discussion

The frequency versus wave-vector dispersion relations for chromium and tantalum along the symmetry directions [100], [110] and [111] may be determined from the solutions of the secular determinant (1) along these directions. Experimental values for the elastic constants have been used in the computation. Those for chromium were taken from Bolef and de Klerk (1963) and those for tantalum from Featherston and Neighbours (1963). The resulting phonon dispersion curves are shown in Figs 1*a* and 1*b* respectively, where we have also plotted the experimental values obtained from neutron scattering.

Experimental phonon dispersion relations for chromium have been obtained by several workers (Møller and Mackintosh 1965; Shaw and Muhlestein 1971). We have compared our theoretical results with those of Shaw and Muhlestein in the three symmetry directions. These experimental curves have not yet been studied on the basis of any of the existing force-constant models. We have compared our theoretical results for tantalum along the three main directions with the dispersion curves obtained by Woods (1964) from slow-neutron scattering experiments.

Figs 1*a* and 1*b* show that there is excellent agreement between the theoretical and experimental dispersion curves for chromium in all three directions but that this is not so in case of tantalum. Along the [110] direction for tantalum, the calculated curves slightly violate the symmetry properties of a body-centred structure. These deviations are attributable to the assumption of short-range interatomic forces, which is incorporated in the Behari–Tripathi model, and to the approximate calculation of the electron–lattice interaction. Analysis of the data by means of the Born and von Kármán theory indicates that the interatomic forces in these metals, particularly in tantalum, are of a fairly long range nature. The incomplete d electronic shells, which characterize these metals and which are responsible for their large cohesive energies (Mott 1962), are also expected to make an important contribution to their lattice dynamical properties. However, as things stand, the present study provides another instance of an adequate interpretation of the lattice vibrations in transition metals by means of the Behari–Tripathi model.

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