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Effects of lattice dispersion and elastic anisotropy on the thermal properties of fcc metals

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Abstract. New parameters which separately take into account the effects of elastic anisotropy and phonon dispersion on the lattice specific heat in the case of fcc metals have been evaluated. A new graded mesh method which uses 162-direction approximation in (1/16) part of the Brillouin zone has been developed to evaluate the two new parameters.

Keywords. Dispersion parameter; anisotropy parameter; Brillouin zone; specific heats.

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1. Introduction

A detailed discussion of the fundamental lattice dynamical models and their application to the study of the thermophysical properties of crystalline solids are available in literature (Joshi and Rajgopal 1968; Rao 1979; Wallace 1972; Maradudin *et al* 1963). In the lattice dynamical model of Debye (1912), the crystal medium is assumed to be dispersionless and that the crystal is elastically isotropic. The experimental neutron diffraction data (Bacon 1962; Dolling and Woods 1965) and other lattice dynamical models (Satya Prakash 1979; Reissland 1972; Brovman and Kagan 1974) show that both the assumptions made by Debye are not obeyed in real crystals.

The anomalous behaviour of θ-T curves gives a qualitative indication of the contribution of the combined effect of dispersion and anisotropy to the lattice specific heat of crystals. The effect of both elastic anisotropy and phonon dispersion have been investigated in the case of cubic crystals (Tolpadi 1979; Singh and Tolpadi 1983). Recently, instead of a θ-T plot a quantitative study has been made regarding the effect of dispersion and anisotropy on the specific heat of some bcc metals (Mohapatra and Tolpadi 1988; Mohapatra 1988).

In the present investigation a 162-direction approximation has been developed to determine quantitatively both the effects of lattice dispersion and anisotropy on the specific heat of some fcc metals. The dispersion and anisotropic parameters α_d and β_a can be defined as (Mohapatra and Tolpadi 1988)

$$\alpha_d = (C_{VM} - C_{VD})/C_{VM}$$
$$\beta_a = (C_{VD} - C_{V\theta})/C_{VM}. \tag{1}$$

Where, C_{VM} is the model specific heat which includes the effects of both phonon

dispersion and lattice anisotropy of the fcc crystal. C_{VD} is the modified Debye specific heat which takes into account lattice anisotropy but excludes the effect of phonon dispersion. $C_{V\theta}$ is the single parameter Debye specific heat which neglects the effects of both phonon dispersion and lattice anisotropy in the crystal. These parameters α_d and β_a have been calculated for Ag, Al, Cu and Ni.

2. Lattice dynamical model

Angular forces were introduced into the lattice dynamics by various workers. In the present study a nine parameter lattice dynamical model has been developed by considering a modified form of angular force which is different from the models of Clark *et al* (1964) and De Launay (1956). The angular force is assumed to be proportional to the change of the angle in the equilibrium plane of the triangles formed by the three atoms. The detailed theory developed for bcc metals has been recently published (Mohapatra and Tolpadi 1988).

In table 1, the types of angles $\theta_1, \theta_2, \theta_3$, etc. and the number of triangles considered in deriving the angular force expression for the fcc crystals are listed up to the fourth nearest neighbours. Using the data given in table 1, the total angular force on a given reference atom can be derived.

The frequency determinant of fcc metals is derived by including the angular force according to the present model. The elements of the dynamical matrix also include the ion electron interaction parameter ak_e as given by Sarkar *et al* (1977) and ion-ion central interaction up to the fourth neighbours. The secular equation of the fcc metals determining the frequencies v of the normal modes of vibration can be written as

$$|D_{ij} - 4\pi^2 M v^2(q) I| = 0 \tag{2}$$

where $i, j = 1, 2, 3$, I is the unit matrix of the order 3, M is the mass of the atom, q is the wave vector and D_{ij} are the matrix elements which includes all the three interactions.

Table 1. Data on the types of angles considered for the calculation of angular forces in fcc crystals.

Neighbourhood	Angular force constant	Types of angles considered	Number of triangles
First	β_1	$\theta_1 = 60°$	24
		$\theta_2 = 90°$	12
		$\theta_3 = 120°$	24
Second	β_2	$\theta_1 = 90°$	8
Third	β_3	$\theta_1 = 48{\cdot}19°$	24
		$\theta_2 = 70{\cdot}53°$	12
		$\theta_3 = 109{\cdot}5°$	12
		$\theta_4 = 131{\cdot}8°$	24
Fourth	β_4	$\theta_1 = 60°$	24
		$\theta_2 = 90°$	12
		$\theta_3 = 120°$	24

The angles do not change when a crystal is subjected to uniform compression (Singh and Dayal 1970; Born 1914; Wilson *et al* 1955; Herzberg 1962). To satisfy this criterion the following additional condition involving the angular force constants is used in the present model

$$(4\beta_1 + (20/3)\beta_2 + 32\beta_3 + 14\beta_4) = 0. \tag{3}$$

3. Evaluation of specific heat and numerical computation

The specific heat of cubic crystals has been investigated by many workers (Tripathi and Behari 1971; Behari and Tripathi 1970; Gupta and Tripathi 1971; Sharma and Awasthi 1979; Shukla and Cavalheiro 1973; Ramamurthy and Satish Kumar 1984) by calculating the phonon frequencies in (1/48) part of the BZ and applying Blackman's sampling technique (Blackman 1935, 1937). At low temperature this method did not reproduce satisfactory results. Singh and Tolpadi (1983) and Mehrotra (1979) had suggested a graded mesh method which uses a 91-direction approximation in (1/16) part of the BZ. It was found that this method did not give the correct values of very low temperature specific heat when $(T/\theta) \simeq 2{\cdot}0 \times 10^{-3}$.

Recently, Mohapatra and Tolpadi (1988) have suggested an accurate method of evaluating the BZ sums of bcc metals by considering a 162-direction approximation in (1/16) of the BZ. In the present study the same numerical technique has been developed to evaluate the specific heat of some fcc metals by taking into account the BZ structure of fcc crystal.

The nine model parameters α_1, α_2, α_3, α_4, β_1, β_2, β_3, β_4 and ak_e that appear in the dynamical matrix elements are evaluated by using the three experimentally known elastic constants C_{11}, C_{12} and C_{44} (Overton and Gaffney 1955; Neighbours and Alers 1958; Alers *et al* 1960; Kamn and Alers 1964) and five frequencies in the principal symmetry directions of the crystal (Svensson and Brockhouse 1967; Birgeneau *et al* 1964; Stedman and Nilsson 1966; Kamitakahara and Brockhouse 1969) along with the condition given in (3). When the secular determinant (2) in the long wave length limit is compared with the elastic constant determinant, the following three relations between the force and the elastic constants are obtained

$$aC_{11} = 2\alpha_1 + 4\alpha_2 + 12\alpha_3 + 8\alpha_4 + 2\beta_1 - 4\beta_2 + \frac{132}{5}\beta_3 + 8\beta_4 + ak_e$$

$$aC_{12} = \alpha_1 + 6\alpha_3 + 4\alpha_4 - 7\beta_1 - 8\beta_2 - \frac{486}{5}\beta_3 - 28\beta_4 + ak_e$$

$$aC_{44} = \alpha_1 + 6\alpha_3 + 4\alpha_4 + \beta_1 + 8\beta_2 - \frac{234}{5}\beta_3 + 4\beta_4. \tag{4}$$

In addition the frequency determinant is solved in some of the symmetry directions of the crystal. The relationship between the force constants and the corresponding frequencies is given below

$$4\pi^2 M v^2 L\,[1\,0\,0] = 8\alpha_1 + \frac{16}{3}\alpha_3 + 8\beta_1 + \frac{1072}{15}\beta_3$$

$$4\pi^2 M v_T^2\,[1\,0\,0] = 4\alpha_1 + \frac{40}{3}\alpha_3 + 4\beta_1 + \frac{664}{15}\beta_3$$

$$4\pi^2 M v^2 L\,[1/2\;1/2\;0] = 6\alpha_1 + 4\alpha_2 + \frac{28}{3}\alpha_3 + 4\alpha_4 - 2\beta_1 + 4\beta_2 + \frac{116}{3}\beta_3 + 4\beta_4 + \frac{1}{2}ak_e$$

$$4\pi^2 M v^2 T_1\,[1/2\;1/2\;0] = 2\alpha_1 + 4\alpha_2 + \frac{20}{3}\alpha_3 + 4\alpha_4 + 10\beta_1 + 4\beta_2 + \frac{124}{3}\beta_3 + 4\beta_4$$

$$4\pi^2 M v^2 T\,[1/2\;1/2\;1/2] = 2\alpha_1 + 4\alpha_2 + \frac{28}{3}\alpha_3 + 10\beta_1 + 12\beta_2 + \frac{116}{3}\beta_3. \qquad (5)$$

These relations are used to evaluate the model parameters uniquely for Cu, Ag, Al and Ni.

4. Results and discussion

In the present investigation, the numerical computation technique described in §3 is used to calculate the thermal properties of fcc metals. Appropriate computer programs have been developed to calculate the force constants, phonon frequencies, the Debye temperature θ_D, specific heats and the parameters α_d and β_a of the four fcc metals viz. Al, Cu, Ag and Ni.

The calculated model specific heat C_{VM} and the phonon dispersion relations in the symmetry directions show very good agreement with the experimental results. The dispersion parameter α_d and the elastic anisotropy parameter β_a defined in (1) are calculated from the computed values of C_{VM}, C_{VD} and $C_{V\theta}$. The temperature variation of α_d and β_a for all the four metals is given in figure 1.

It is observed that α_d is positive in the entire range of temperature for all the four fcc metals. In the low temperature range when $(T/\theta) \leqslant 0{\cdot}01$ the effect of dispersion on lattice specific heat is negligible. In this low temperature region the Debye T^3 law is valid. In the higher temperature region with $(T/\theta) \geqslant 1{\cdot}5$, α_d and β_a are negligible so that C_V attains the classical value. α_d is positive in the entire range of temperature and negligible in the low and high temperature regions. Therefore, the temperature variation of α_d is expected to show a maximum in the intermediate temperature range. The maximum values of dispersion parameter α_d are given in table 2. α_d shows a maximum when $T/\theta \simeq 0{\cdot}13$. In the case of aluminium which is nearly elastically isotropic ($s = 0{\cdot}84$) the effect of dispersion is less as compared to other fcc metals.

The temperature variation of the elastic anisotropy parameter β_a given in figure 1 is found to be negative in the entire temperature range. It is negligible in the low and high temperature regions for all the cases. Further β_a shows a minimum when $(T/\theta) \simeq 0{\cdot}23$. The minimum values of β_a given in table 2 show that the magnitude of β_a is minimum for aluminium ($s = 0{\cdot}84$). The temperature variation of the parameter $(\alpha_d + \beta_a)$ for Cu, Ag, Al and Ni is plotted in figure 2. These curves represent the

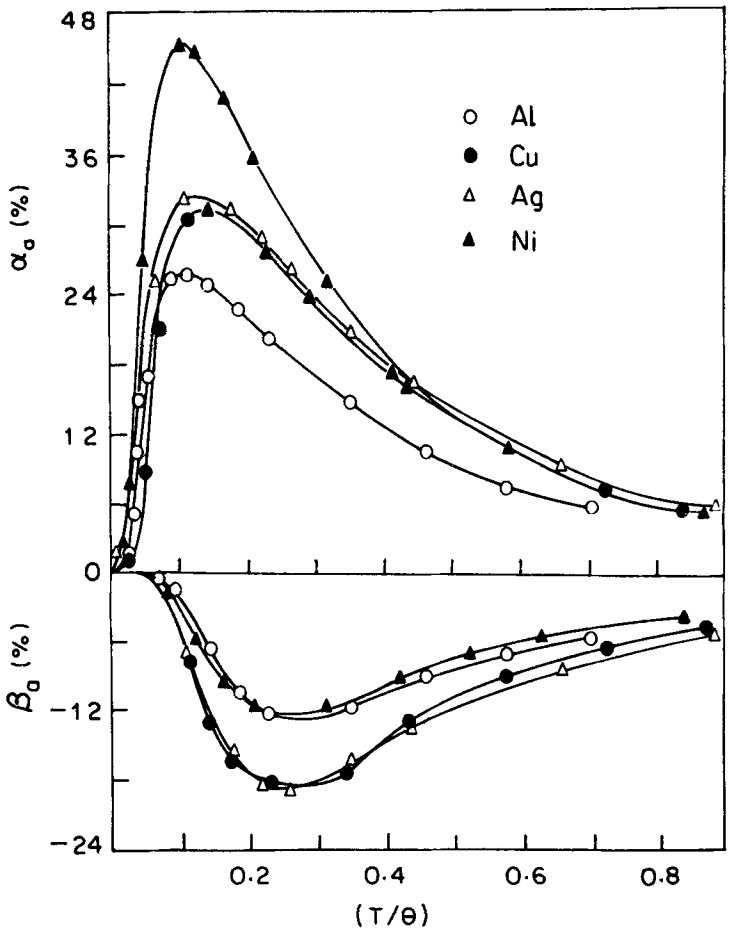

Figure 1. Temperature variation of dispersion parameter (α_d) and anisotropic parameter (β_a) for fcc metals Ag, Al, Cu and Ni.

Table 2. Maximum dispersion parameter (α_d) and minimum anisotropic parameter (β_a) of some fcc metals.

Metal	Anisotropy factors	Temperature variation of α_d and β_a			
		α_d	(T/θ)	β_a	(T/θ)
Cu	0·313	31·5	0·14	−17·8	0·23
Ag	0·334	32·0	0·11	−18·3	0·26
Ni	0·42	45·3	0·10	−11·5	0·21
Al	0·843	25·9	0·11	−12·0	0·23

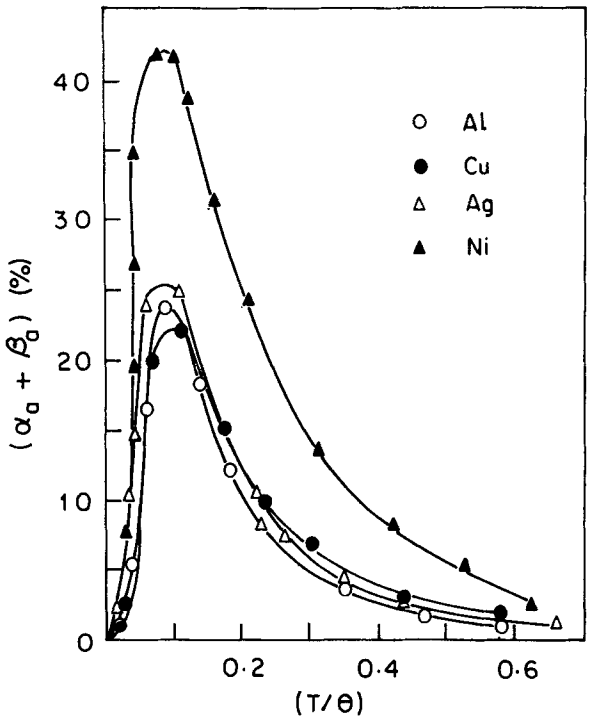

Figure 2. Temperature variation of $(\alpha_d + \beta_a)$ for fcc metals Ag, Al, Cu and Ni.

departure from the Debye criterion. In bcc metals Na and K (Mohapatra and Tolpadi 1988) $(\alpha_d + \beta_a)$ is positive when (T/θ) varies from 0·01 to 0·12 and it is negative when $(T/\theta) \geqslant 0{\cdot}12$. But in fcc metals it is found that $(\alpha_d + \beta_a)$ is positive throughout the entire range of temperature region. The combined effect of dispersion and anisotropy on the specific heat of fcc metals Al, Cu, Ag and Ni shows a maximum when $T/\theta \simeq 0{\cdot}1$.

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