Three-body interactions, rotational invariance and the elastic consistency of lattice dynamical models for face-centred tetragonal indium

V RAMAMURTHY and S B RAJENDRAPRASAD*
Department of Physics, Indian Institute of Technology, New Delhi 110016
*Present address: Advanced Centre for Material Sciences, Indian Institute of Technology, Kanpur 208016, India

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Abstract. The origin of the elastic inconsistency of DAF, MAS and GTF models for non-cubic solids and the failure of their force constants to comply with all the rotational invariance conditions are analysed by resolving the atomic displacements of face-centred tetragonal indium along three mutually perpendicular directions. It is shown that a lattice dynamical model suffers from these deficiencies as a consequence of its neglect of three-body interactions as well as the mixed neighbour interactions associated with the angular forces, while the CGW model which incorporates both these interactions is elastically consistent and its potential energy rotationally invariant. The degree of equivalence that exists among the force constants of DAF, MAS, GTF and CGW models, the distortions introduced by the elastic inconsistency into the phonon dispersion curves of fct indium as well as the consequences of imposing the rotational invariance conditions on the force constants of a lattice dynamical model are discussed.

Keywords. Elastic inconsistency; phonon dispersion curves; rotational invariance; three-body interactions; CGW model; fct indium.

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1. Introduction

Irrespective of the differences in their assumptions regarding the nature and range of interatomic forces, most of the lattice dynamical models proposed so far (Begbie and Born 1947; De Launay 1956; Lehman et al 1962; Rathore and Verma 1977) are not at all invariant with respect to rigid body rotations even in the case of high symmetry cubic crystals. The harmonic force constants of these models comply with the Born-Huang conditions (Born and Huang 1954) so long as the crystal is in equilibrium, but they fail to satisfy the additional conditions which reduce the change in potential energy to zero (Gazis and Wallis 1966; Keating 1966). Besides, none of these models incorporates either the three-body interactions (Martin 1975) or the mixed neighbour interactions (Rajendraprasad 1982). Nevertheless all models are elastically consistent and give rise to unique force constant expressions for elastic constants as a consequence of the cubic symmetry. However, the transition from cubic to tetragonal or hexagonal symmetry destroys the equivalence between x (or y) and z directions and invariably increases the number of force constants that appear in the elements of the dynamical matrix, $D(q)$. Further, the apparent differences in the expressions for $C_{44}$, obtained by the method of
long waves from $D_{zz}$ and $D_{zz}$ elements of fct indium (see table A in appendix), demonstrate unambiguously the elastic inconsistency of various force constant models. Hence all those lattice dynamical studies of non-cubic metals based on these inadequate models (Slutsky and Garland 1957; Slutsky and Livingston 1960; Wolfram et al 1963; De Wames and Lehman 1964; De Wames et al 1965; Ashokkumar 1973; Sharma and Upadhyaya 1977; Ramamurthy and Rajendraprasad 1981) are devoid of any physical significance. Perhaps this is one of the reasons why none of the force constant models was used to investigate the lattice dynamics of fct indium.

It is therefore essential to ascertain the basic factors responsible for the elastic inconsistency of a force constant model which are completely obscured by the cubic symmetry and to develop elastically consistent models for the lattice dynamical study of non-cubic metals. Keeping these twin objectives in mind, the present authors formulated a variety of force constant models for fct indium. Besides recognizing its elastic consistency, these authors made use of CGW model (Clark et al 1964) for the first time, to deduce the phonon dispersion curves along the principal symmetry directions as well as the phonon frequency distribution function of fct indium (Ramamurthy and Rajendraprasad 1983, hereafter referred to as I) which are in very good agreement with the corresponding experimental results (W Reichardt and H G Smith, unpublished, private communication) and pseudopotential calculations of Garrett and Swihart (1976). Despite the fact that this model for fct indium is rotationally invariant, it is not at all clear whether there is any direct link between these two deficiencies of other models. Further, Ramamurthy (1982, hereinafter referred to as II) has shown that the CGW model is not equivalent to DAF model in the case of cubic crystals by analysing the fundamental differences between them. The present authors have therefore extended this analysis to CGW, DAF, MAS and GTF models for fct indium to determine the nature of the interatomic forces responsible for the elastic inconsistency and to ascertain the manner in which the CGW model gets rid of it. It is the purpose of this paper to describe these investigations which reveal the importance of three-body forces and mixed neighbour interactions while the role of the rotational invariance conditions in the lattice dynamics of non-cubic crystals is described in a subsequent paper.

2. Theory

To facilitate the analysis of various force constant models, the dynamical matrix, $D(q)$ is split into an ionic part, $D^i$ and an electronic part, $D^e$ representing contributions from short range ion-ion interactions and long range electron-ion interactions, respectively. An electron gas model which satisfies the symmetry requirements of the lattice (Ramamurthy and Singh 1978) was used to express the latter and the elements of $D^e$ which include the contributions from normal and umklapp processes are given by equation (2.5) in I. In order to express the elements of $D^i$ in terms of central and angular, axially symmetric or general tensor forces, we follow the procedure adopted by Ramamurthy (1982) and consider three atoms at $O$, $A$ and $B$ in figure 1 which form a triangle with angle $\theta$ at the origin. The change in the potential energy of the atom at $O$ due to displacements $s_o$, $s_A$ and $s_B$ of these atoms from their equilibrium positions, in the harmonic approximation, is given by

$$dV = \frac{1}{2} \beta \left[ (\hat{\rho}_A \cdot (s_A - s_o))^2 + (\hat{\rho}_B \cdot (s_B - s_o))^2 \right] + \frac{1}{2} \eta \left[ (d\phi_A)^2 + (d\phi_B)^2 \right]$$

$$+ \frac{1}{2} \delta (d\theta)^2,$$  \hspace{1cm} (1)