Analysis of Phonon Dispersion Relations
From Atomic Model to Continuum Theory

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Abstract

The phonon dispersion relations calculated by atomic model: shell model and ab initio, and by microcontinuum theory are introduced in this paper. It is found that both atomic models and microcontinuum theory can describe the common feature of phonon dispersion relation for the crystal having more than one atom per primitive cell and can give a good fit to the experimentally measured data. The optical branches are absent from the classical continuum theory and the strain gradient theory due to the neglecting of the atomic structure of the crystal. The importance of phonon dispersion relation and the applicability of microcontinuum theory are discussed.

Keywords: Phonon dispersion relation, Microcontinuum theory, Atomic model, Lattice dynamics

1. Introduction

The dynamics of atoms in crystals - lattice dynamics - is basic to many fields of study in the solid state. It turned out to be most readily described not in terms of the individual atoms, but in terms of travelling waves, named lattice vibrations by Born [1], each characterized by a wave vector, a frequency, and certain polarization properties. These waves are the normal modes of vibration of the system. The quantum of energy in an elastic wave is called a phonon. At a very low temperature a solid can be regarded as a volume containing non-interacting phonons. The vibrating lattice can be considered as a set of phonon excitations. Physical properties of solid, including the thermal, optical and electrical properties, are directly dependent upon the frequency-wave vector relationship, which is called the phonon dispersion relation. Phonon dispersion relation is thus an essential feature for the dynamics of atoms in crystal and is of special importance for solid state physics and material science.

Phonon dispersion relations can be calculated from atomic force by the discrete atomic model. The analog of atomic force is the constitutive relation in the continuum theory, which serves to distinguish one material from another. It follows that the phonon dispersion relations can be also calculated through the constitutive relations of a material by a continuum model if the model can manifest the dynamics of atoms in crystals.

Due to the availability of both computational and experimental determination, phonon dispersion relations have dual significance: to understand the excitations that crystal lattice can experience and their consequences for the thermal, optical and electric properties, and to determine the atomic force constants from the measurement of phonon dispersion relations. For microcontinuum theory, phonon dispersion relations can provide a basis for experimental observations to determine the micromorphic elastic constants and to compare with the predications of atomic lattice dynamics so as to examine the validity and applicability.

Single crystal silicon is the most important material in MEMS technology. In this paper, it is taken as a material example.

2. Phonon Dispersion Relations Based on Atomic models

There are two levels to calculate the phonon dispersion relations: the phenomenological level, or the *ab initio* level.

In the phenomenological level, the theoretical models which have been employed for constructing the dynamical matrix for tetrahedrally bonded crystals with diamond and zincblende structures can be broadly classified in three categories: (i) force constant models, (ii) the rigid ion model and (iii) the shell models.

In *Born-von Karman force constant model*, by assuming periodic boundary conditions and small displacements, the expression for the potential energy of the crystal was given as

$$\Psi_2 = \frac{1}{2} \sum_{k,l=1}^{n} \sum_{\alpha,\beta=1}^{\nu} \sum_{i,j=1}^{3} \phi_{ij}(k\alpha, l\beta) u_i(k\alpha) u_j(l\beta) , \qquad (1)$$

where index, k, or l, identifies a unit cell with ν atoms, α or β a particular atom in a unit cell. The equation of motion is

$$m_{k\alpha}\left(\frac{\partial^2}{\partial t^2}u_i(k\alpha)\right) = -\sum_{l=1}^{n}\sum_{\beta=1}^{v}\sum_{j=1}^{3}\phi_{ij}(k\alpha, l\beta)u_j(l\beta) \quad . \ (2)$$

Given a wave vector q, the displacement of an atom can be written as

$$\qquad\qquad , \quad (3)$$

and the equation of motion then becomes

$$\qquad\qquad , \quad (4)$$

where M is called the dynamical matrix, and it reads

$$\qquad\qquad . \quad (5)$$

Only 2 parameters are needed to describe nearest-neighbour interactions. However, it was found that 15 independent force constants extending to fifth nearest neighbours were required to give a good fit to the phonon frequencies measured by neutron spectroscopy [2].

In the *rigid ion model*, the forces in the crystal arise from two contributions: (a) long-range or Coulomb interactions between 'effective' charges on the ions, and (b) short-range central and non-central interactions, as described in the Born-von Karman force constant model. For a good agreement with the experimental phonon dispersion curves, usually 11 adjustable parameters are needed [3].

In the *shell model*, an atom is represented by a non-polarisable ion core and a shell of valence electrons. The displacement of the shell relative to the core produces a dipole. An ion core and its shell are regarded as point charges when dealing with the Coulomb interaction with other ion cores and shells. The energy of the crystal can be given in the harmonic approximation in the following form

$$\qquad\qquad , \quad (6)$$

where $p(k\alpha)$ is the electronic dipole moment of an ion, and $E(k\alpha)$ is the effective field at the site $(k\alpha)$. Terms not involving the dipole moments give the energy of rigid ion model, referred to as the short-range interaction and Coulomb interaction. The other term are introduced to take account of the energy of the dipoles which arises from their

short-range interactions with one another and with displaced ions, and their energy of interaction with the field. Five independent parameters are needed to describe core-core, core-shell and shell-shell forces extending only to nearest neighbours. For a successful explanation of phonon dispersion in many diamond/zincblend type semiconductors it is necessary to use a 14-parameter model [3].

The quantum mechanical calculation of phonon spectra began in the early 1970s, which has drawn a lot interest towards first-principle studies of lattice dynamics of semiconductors and insulators. An *ab initio* calculation requires an accurate and parameterless knowledge of the microscopic electronic response to frozen-in lattice vibrations. The basic idea is to determine force constants via the total energy of the crystal under investigation. Within the adiabatic approximation, the total energy is a function of only the position of the ions, and can be divided into the direct ion-ion interaction energy and the energy of the valence electrons moving in the potential field of ions, including quantum mechanical kinetic, exchange and correlation energies. There are two commonly used approaches for the calculation of the total energy: (i) a direct approach and (ii) a linear response approach.

The phonon dispersion relation, i.e., the ω - q relations, calculated based on a shell model [4], an *ab initio* model [5], and measured from neutrons scattering, for silicon, are shown in Fig.1.
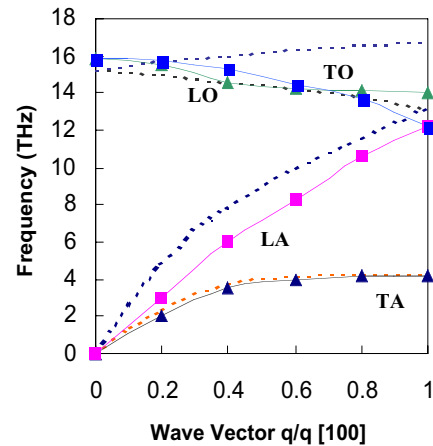

Fig.1 Phonon dispersion relations
solid line--shell model [4], dot line--*ab initio* [5],
points--experimental measurement [4]

3. Phonon Dispersion Relations Based on Microcontinuum Theory

Classic continuum mechanics views the crystal as a homogeneous continuous medium rather than as a periodic array of atoms. The continuum approximation is usually valid for elastic waves of wavelengths longer than 10^{-6} cm,

which means for frequencies below 10^{11} or 10^{12} Hz. Assuming a harmonic wave, classical continuum mechanics yields a non-dispersive acoustic waves with constant speeds for longitudinal and transverse waves moving along [100], respectively, as

$$\qquad , \qquad , \qquad (7)$$

where ρ is the density, C_{11}, C_{12}, C_{44} are the elastic constants.

The gradient-dependent plasticity theory is an extension of classic continuum theory to the application in micron range. It incorporates a plastic strain gradient into the constitutive relations. The wave equation for one-demensional gradient-dependent plasticity theory was derived by de Borst et al [6]. For a single harmonic wave propagating though one-dimensional element, the gradient-dependent plasticity theory yields a dispersion relation for acoustic wave with a frequency

$$\omega = K\sqrt{E/\rho}\sqrt{(h+gK^2)/(E+h+gK^2)}, \qquad (8)$$

where h is the hardening/softening modulus, E the Young's modulus, g is a coefficient in the Taylor series expansion of the yield function.

Microcontinuum field theory, micromophic theory, developed by Eringen [7] constitutes extensions of the classical field theories concerned with the deformations, motions, and electromagnetic (E-M) interactions of material media, as continua, in microscopic space and short time scales. In terms of a physical picture, a material body is envisioned as a collection of a large number of deformable particles, each particle possesses finite size and direction representing its microstructure. The particle has the independent degrees of freedom for both stretches and rotations, in addition to the classical translation degrees of freedom of the center, and may be considered as a polyatomic molecule, a primitive unit cell of a crystalline solid, or a chopped fiber in a composite, etc.

In the absence of external force, the equations of motion for micromorphic material can be written as

$$t_{kl,k} = \rho\ddot{u}_l \qquad , \qquad (9)$$

$$m_{klm,k} + t_{ml} - s_{ml} = \rho j \ddot{\phi}_{lm} \qquad , \qquad (10)$$

where t is the Cauchy stress, m the moment stress, and s is microstress average. The constitutive relations for isotropic micromorphic solid can be written as

$$\qquad , \qquad (11)$$

$$\qquad , \qquad (12)$$

$$\qquad , \qquad (13)$$

where $\qquad$ are the 16 material constants, the strain measures are related to the macro- and micro-motions, u and ϕ, as

$$\qquad , \qquad (14)$$

$$\qquad - \qquad , \qquad (15)$$

$$\lambda_{ijk} = \phi_{jk,i} \qquad . \qquad (16)$$

Assuming harmonic waves

$$\qquad (17)$$

$$\qquad (18)$$

propagating in the [100] direction, 3 acoustic and 9 optical waves can be obtained and grouped as

(1) RO (rotational optical wave)

$$\qquad (19)$$

(2) SO (shear optical waves)

$$\qquad - \qquad (20)$$

(3) LA, LO, LDO (longitudinal acoustic, optical and dilatational optical waves)

$$\qquad (21)$$

(4) TA, TO, TRO (transverse acoustic, optical and rotational optical waves)

$$\qquad (22)$$

where, $\qquad$ are related to the 16 material constants, and

$$\frac{\omega}{?} \quad - \qquad , \qquad (23)$$

$$\frac{\omega}{?} \quad - \qquad , \qquad (24)$$

$$\overset{\vee}{?} \quad - \qquad , \qquad (25)$$

$$\overline{?} \; - \; \qquad \qquad . \qquad (26)$$

The material constants in the microcontinuum theory for single crystal silicon are determined by fitting the phonon dispersion relations to the experimental measurements. The results for LA, TA, LO, TO branches are shown in Fig.2.
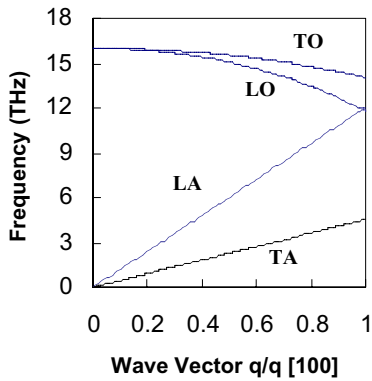

Fig.2 Phonon dispersion relations
based on microcontinuum theory

4 Discussions

Crystalline solids are distinguished from other states of matter by a periodic arrangement of the atoms. With crystal having more than one atom per primitive cell, for example, silicon, the dispersion relation ω versus K develops two branches, known as the acoustic and optical branches. The very long waves of acoustic branch are in fact identical with the elastic vibrations with the atoms in the same cell moving practically in unison as a rigid unit. These are external modes. While the wave of optical branches are internal modes involving stretching of the covalent bonds (longitudinal) and the distortion of the cells (transverse). They are higher frequency modes. Highest frequency modes may involve the rotations, coupling of the dilatation and rotations.

All phenomenological theories of lattice dynamics can give a good fit to the experimentally measured phonon frequencies. However, their parameters do not contain conceptual simplicity. That is one reason why a *priori* quantum mechanical calculation of phonon spectra was needed and developed. On the other hand, microcontinuum theory treats the material as a continuum, dramatically enlarge the region of applicability, and whereas it can describe the essential feature of the phonon dispersion relations and give good fit to the experimental measurements.

The optical vibrations of long wavelengths are of special importance in considering optical and electrical behavior of crystal. Under an electromagnetic field it is the optical modes that are excited. Optics is a phenomenon that necessitates the presence of an electromagnetic field. In general an electromagnetic wave interacts only with lattice vibrations of the same wavelength, and will be strongly affected only if its frequency is near that of the latter. The frequencies of lattice vibrations are generally in the range from 0 to   Hz, hence light waves of similar frequencies have wave-lengths larger than   = 0.003cm (c, the speed of light), which are enormously large compared with the lattice constants of crystal. Therefore the lattice vibrations that can interact appreciably with light are very long waves. In the long optical vibrations, the opposed motions of the oppositely charged particles give rise to a net oscillating dipole moment of a cell, which is totally absent in the long waves of acoustic branch, where the opposite charges move in unison. Therefore only the long waves of the optical branch will concern us in the discussion of the optical and electrical behavior of crystals [8].

Since optical branches are internal vibration modes, the absence of optical branch is due to the neglecting of the atomic structure of crystal. From this viewpoint, classical continuum theory, gradient theory would break down if the micromotion and the microstructure of material become important for an object under investigation.

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