Model-potential approach to the lattice dynamics of barium

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A simple form of a model potential has been used to investigate the phonon spectra of barium. The present model-potential approach reproduces the result that the frequencies of the transverse branches are higher than those of the longitudinal branches in the [100] direction. The results obtained show reasonable agreement with the experiment for all but the [110] direction.

Recently, Mizuki, Chen, Ho, and Stassis[1] presented inelastic neutron scattering results for the phonon dispersion of the alkaline-earth-metal barium. The general features of the electronic band structure of the alkaline-earth metals are found to be in between those of the simple and transition metals. In fact, it has been found that below the Fermi level, the electronic bands are almost free-electron-like.[2-5] This leads us to believe that the pseudopotential approach is applicable to these metals. Furthermore, good agreement for calcium was recently obtained by Ambika Prasad, Gupta, and Tripathi[6] using the model-potential approach. Band-structure calculations also indicated that the electronic wave functions at the Fermi level contain a substantial admixture of d character arising from hybridization with the d bands, which in these metals are slightly above the Fermi level. We[7-9] have been able to successfully explain the phonon dispersion of d-band-characterized noble and transition metals within a pseudopotential framework. In this paper, we try to investigate the phonon dispersion in bcc barium. We obtain good agreement between theory and experiment. Particularly, the frequencies of the longitudinal branches have been found to be lower than those of the transverse branches in the [100] direction as has been observed.

The dynamical matrix elements $D_{\alpha\beta}(\mathbf{q})$ are composed of two parts, namely, the electrostatic contribution $D^{C}_{\alpha\beta}(\mathbf{q})$ and the electronic contribution $D^{E}_{\alpha\beta}(\mathbf{q})$. The electrostatic contribution to the dynamical matrix elements arises from the direct Coulomb interaction between the ions immersed in a uniform compensating negative charge and can be evaluated on the lines of Ref. 10.

The electronic contribution to the dynamical matrix is calculated using the following expression:[6-9]

$$D^{E}_{\alpha\beta}(\mathbf{q}) = 2\sum_{\mathbf{h}}{}'[(q_\alpha + h_\alpha)(q_\beta + h_\beta)F(|\mathbf{q}+\mathbf{h}|) - h_\alpha h_\beta F(|\mathbf{h}|)] \ , \tag{1}$$

where $\mathbf{h}$ are reciprocal-lattice vectors and $F(q)$ is the energy–wave-number characteristic given by

$$F(q) = -\frac{\Omega_0 q^2}{8\pi z e^2}|\omega_b(q)|^2 \frac{\epsilon^*(q)-1}{[1-G(q)]\epsilon^*(q)} \ . \tag{2}$$

Here z is the valency of the ion, e the electron charge, Ω_0 the atomic volume, and $\omega_b(q)$ the bare-electron–ion model potential as given by Nand, Tripathi, and Gupta.[9] The dielectric function $\epsilon^*(q)$ is given by Moriarty[11] as

$$\epsilon^*(q) = \epsilon(q) - G(q)[\epsilon(q)-1] \ , \tag{3}$$

where $\epsilon(q)$ is the Hartree dielectric function and $G(q)$ is the local field correction due to Singwi, Tosi, Land, and Sjolander.[12] To calculate the electronic contribution to the dynamical matrix, the parameters of the model potential were taken as $r_1 = 1.26$ a.u. and $r_2 = 2.532$ a.u., where r_2 is the ionic radius of barium, taken from Weast,[13] and r_1 the core radius which is adjusted to fit the zone-boundary frequency in the [100] direction. The convergence in the summation of the electronic contribution to the dynamical matrix was obtained after summing over 1198 reciprocal-lattice vectors up to $\sqrt{70}(2\pi/a)$.

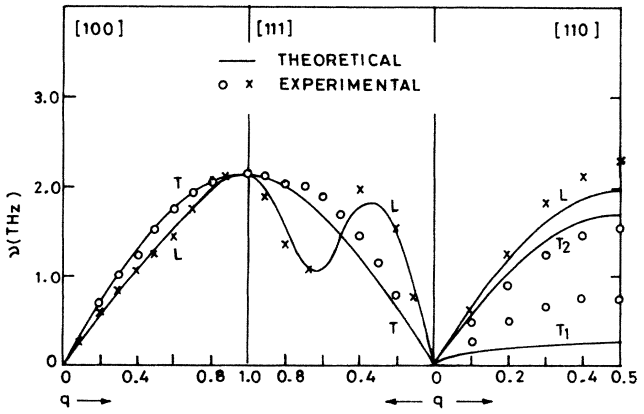

FIG. 1. Phonon dispersion curves of bcc Ba.

Our calculated values for the phonon frequencies for barium in the three symmetry directions are shown in Fig. 1. As is obvious from the figure, a reasonable agreement has been obtained between theory and experiment, the maximum discrepancy being of the order of 10%–15%, except for the [110] T_1 branch. In particular, we obtain frequencies of the longitudinal dispersion curve which are lower than those of the transverse branch along the [100] symmetry direction, a surprising result which was also obtained experimentally. It should be mentioned that the phonon frequencies, calculated by Animalu[14] and Sharma[15] using local pseudopotentials, are higher by an order of 2 when compared with the experiment. Even the phonon frequencies calculated by Moriarty[16] using the generalized pseudopotential approach deviate by 40% as compared to the measured values. Furthermore, these earlier calculations do not produce frequencies of the [100] transverse dispersion curve higher than those of the longitudinal branch.

We conclude that the present model-potential treatment has been successful in explaining the lattice-dynamical properties of alkaline-earth metals.

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