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Lattice dynamics of lanthanides

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Résumé. — Les fréquences de phonons des deux lanthanides Tb et Ho ont été calculées à partir du modèle de potentiel non local des métaux de terres rares d'Upadhyaya et Animalu, qui prend en compte les termes linéaires en E et $\mathbf{k} \cdot \mathbf{q}$ dans le développement de Taylor du facteur de forme. Les termes du deuxième ordre en E ne contribuent pas aux expressions des fréquences de phonons. L'importance des forces non binaires dans la dynamique du réseau de Tb et Ho a également été considérée.

Abstract. — Phonon frequencies of two lanthanides, namely Tb and Ho, have been calculated by applying the rare earth metal model potential of Upadhyaya and Animalu in a nonlocal scheme which accounts the linear E - and $\mathbf{k} \cdot \mathbf{q}$ -dependent terms in the Taylor's expansion of the form factor. Second order E -dependent terms in the expansion do not contribute to the phonon frequencies. Importance of unpaired forces in the lattice dynamics of Tb and Ho has been also discussed.

The lanthanide group of rare earth metals is characterized with the outer electronic configuration $4f^n 5d^m 6s^2$ ($n = 2, 3, \dots, 14$; $m = 1, 2$) and covers the elements with atomic numbers from $Z = 58(\text{Ce})$ to $Z = 71(\text{Lu})$. In this group of metals, the ions are predominantly trivalent and the term values vary only slightly over the series. Recently Upadhyaya and Animalu [1] have developed a model potential of Heine-Abarenkov (HA) type to represent the electron-phonon interaction in rare earth metals.

Similar to the transition metal model potential (TMMP) method of Animalu [2] in which the s-d hybridization is incorporated through an $l = 2$ model potential well depth, the s-f hybridization in the energy bands of the lanthanides in the REMMP is characterized by an $l = 3$ model potential well depth of the resonant form $A_3(E) \propto \frac{1}{2} W_f / (E_f - E)$, where E_f is the position and W_f the width of the f-band resonance. The REMMP has been applied by Upadhyaya and Animalu [1] in the calculation of phonon dispersion in two lanthanides, namely Tb and Ho, by including the nonlocality in the Eschrig and Wonn scheme [3] and a reasonably good agreement is found between the computed and experimental results. In this scheme [3], the form factor is expanded in terms of E - and $\mathbf{k} \cdot \mathbf{q}$ -dependent terms in Taylor's series and the expansion is truncated upto linear terms. The expressions for the coefficients of E - and $\mathbf{k} \cdot \mathbf{q}$ -dependent terms have been derived by Upadhyaya and Animalu [1], and Upadhyaya [4]. If the full scheme of Eschrig and Wonn [3] together with a relation of the coefficients of E - and $\mathbf{k} \cdot \mathbf{q}$ -dependent terms of refe-

rence [4] is applied in the calculation of phonon frequencies of Tb, it is observed that the phonon spectrum is raised in general by 10 to 15 % and a similar fit to that of Upadhyaya and Animalu [1] is obtained for a bit lower value of the nonlocal parameter a_q . Next, it was thought that second order E -dependent terms may be contributing substantially, but their inclusion does not affect the phonon frequencies of Tb and Ho.

Standard procedure has been adopted for the phonon frequency calculation of the hcp system. The Coulombic contribution to the dynamical matrix has been calculated as usual and the repulsive contribution has been assumed to be negligible, because (i) of the parametrized treatment of the nonlocality in the present work and (ii) the hard core repulsion may be cancelling in rare earth metals by the f-f attraction similar to d-d attraction in transition metals [5]. The band structure contribution to the dynamical matrix involves an expression for the energy wave number characteristic $G(q)$ whose expression is obtained by including the first and second order E -dependent terms and first order $\mathbf{k} \cdot \mathbf{q}$ -dependent term in the Taylor expansion of the model potential as following :

$$G(q) = G_0(q) \left[1 + a_q \frac{C(q)}{D(q)} + b_q \frac{E(q)}{D(q)} + c_q \frac{F(q)}{D(q)} \right] \quad (1)$$

where $G_0(q)$ is the energy wave number characteristic in the local approximation,

$$a_q = \frac{1}{V^{\text{loc}}(q)} \cdot \left(\frac{\hbar^2}{2 m^*} \right) \left(\frac{\partial F}{\partial E} \right)_{E_F}, \quad b_q = \frac{2 k_F}{\sqrt{3} q} \cdot a_q$$

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and

$$c_q = a_q^2 + \frac{1}{2} (\hbar^2/2m^*)^2 \frac{1}{V^{\text{loc}}(q)} \cdot \left(\frac{\partial^2 F}{\partial E^2} \right)_{E_F}$$

The expressions for $C(q)$, $D(q)$, $E(q)$ and $F(q)$ are the following :

$$\begin{aligned} C(q) &= \frac{3}{2} Z \left(1 + \eta^2 - \frac{(1 - \eta^2)^2}{2\eta} \ln \left| \frac{1 + \eta}{1 - \eta} \right| \right), \\ D(q) &= \frac{3}{2} \frac{Z}{k_F^2} \left(1 + \frac{1 - \eta^2}{2\eta} \ln \left| \frac{1 + \eta}{1 - \eta} \right| \right), \\ E(q) &= \theta(\eta - 1) 4 k_F^2 \eta(1 - \eta) D(q), \\ \text{and} \\ F(q) &= \frac{3}{2} Z \frac{k_F^2}{3} \times \\ &\quad \times \left[1 - \frac{40}{3} \eta^2 - \eta^4 - \frac{(1 - \eta^2)^3}{2\eta} \ln \left| \frac{1 + \eta}{1 - \eta} \right| \right] \end{aligned} \quad (2)$$

where $\eta = q/2k_F$ and θ is the Heaviside function.

In the present treatment, we take a_q , say a , as an adjustable parameter because of the scarcity of the spectroscopic data, needed for their evaluation and compute the phonon dispersion in Tb along ΓA , ΓM and ΓKM symmetry directions with $a = -1$. The results of calculation are shown in figure 1 by the dot-dash curves (only along ΓA direction) with the experimental points of Nicklow *et al.* [6] by considering the first three terms of $G(q)$ in eq. (1). The phonon spectrum is seen to be raised by about 10 to 15% when compared to the results of Upadhyaya and Animalu [1] which has been shown by dashed lines in figure 1. To obtain a better fit, we find that $a = -0.85$ gives the phonon frequencies of Tb of the same order as in reference [1] and present the dispersion curves by solid lines. It is to be seen that now the curves are indistinguishable to that of Upadhyaya and Animalu [1] along ΓA direction but the curves are improved along ΓM direction. Next, the calculations were also done by including 4th term of $G(q)$ in eq. (1), but no effect was found on the phonon frequencies of Tb. Similar results are obtained for Ho. We expect the similar conclusions for other hcp lanthanides.

Spin-orbit coupling is also important for the lanthanides, having high atomic number. Upadhyaya and Animalu's calculations [1] show that the contribution due to this factor is not substantial as far as the question of phonons in lanthanides is concerned. Local field corrections seem also important in the

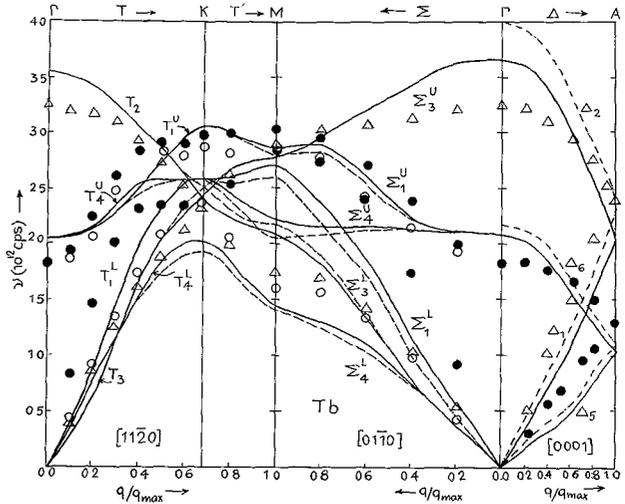


Fig. 1. — Phonon dispersion in Tb : — solid curves represent the calculations with $a = -0.85$, .-. dot dash curves with $a = -0.1$ and - - - dashed curves are those of Upadhyaya and Animalu [1]; Δ \bullet \circ are the experimental points.

rare earth metals. This can be done by following the method as discussed by Upadhyaya and Animalu [1].

It is to be mentioned that in the present calculations, only central paired interaction is involved and hence the ordering of dispersion branches along ΓKM direction and the desired degeneracy at the K-point in Tb and Ho cannot be explained [7]. This can be explained by accounting third order terms in the pseudopotential similar to Bertoni *et al.* [8]. The inclusion of local field effects gives rise to multi-ion forces and hence may explain the desired results of ΓKM direction. We have not done these complex first principles calculations because of the large computational effort and time involved in this work. However, we have developed a force constant approach which accounts for the three body forces in a simple way and explains the ordering of dispersion curves along ΓKM direction in Tb and Ho [9]. This procedure does not include for the electron gas compressibility and hence a force constant approach, consistent with the recent work of Upadhyaya [10] for the hcp system will be satisfactory.

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