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A MODEL POTENTIAL TO STUDY THE LATTICE DYNAMICAL PROPERTIES OF METALS

by

Anand Swaroop

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In Loving Memory of my Mother

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PREFACE

The theoretical studies of the lattice dynamical properties of solids have been of immense importance for the past twenty years or so. Recent developments in the experimental techniques to study these properties have opened new avenues for theoretical research. Side by side more accurate theories based on much more sound footing and having least approximations have been developed to interpret the experimental data. Particularly, the case of metals has been very interesting because of the presence of conduction electrons which give rise to the electron-ion interaction. It is a many body interaction characterized by long range screening of ions by electrons. The treatment of the same has been problematic in the past. The phenomenological theories[†] have been developed to incorporate it explicitly, but these all have met with limited success. During the past decade or so, pseudopotential theory[2] has emerged as one of the most simple and most powerful technique to study the electron-ion interaction. This pseudopotential model has also been applied quite successfully in interpreting atomic, electronic and numerous other lattice dynamical properties of metals. Cohen and Heine[3] have summarized the empirical model potentials which have been obtained for a large number of elements.

In the present investigation the author has suggested a different way to improve upon the delta function approximation which is used by Harrison in constructing the point-ion model potential[2]. It has been accomplished by replacing the delta function by gaussian one and combining it with $-ze^2/r$ term a new form of potential containing two adjustable parameters has been proposed. It is apparent from its form that the corresponding matrix elements of gaussian function approach zero value at large q as demanded by full theory. To include the effect of screening the bare ion form factor has been divided by a suitable dielectric function. Recent studies[4] show that the Hartree dielectric function incorporating exchange and correlation effects according to Singwi et al.[5] gives quite reasonable treatment for screening. The same dielectric function has been employed throughout in the present study. The screened form factors of matrix elements thus obtained have been applied to study the various properties of all the alkali metals but cesium as no experimental results on phonon dispersion for this particular metal are available.

Thus the present work comprises of the theoretical studies on the lattice dynamical properties of alkali metals except cesium using the proposed model potential. Besides this the author has also studied the lattice dynamics and thermal properties of gold using a semiphenomenological model where the interaction has been treated in a realistic manner. Among the noble metals particularly the case of gold has been found to be more interesting because of the anomalous behaviour of its θ_D versus T curve at low temperatures[6]. The entire work is being presented in seven chapters. A chapterwise summary is given. below.

In first chapter the excerpts from pseudopotential theory along with a review of different forms of pseudo-potentials have been presented. The simplicity and wide applicability of the pseudopotential approach have also been discussed.

The second chapter deals with the calculation of form factors, energy wave number characteristics and Cochran's G(q) function. The Fourier transform of unscreened model potential has been taken which is further divided by a Hartree dielectric function modified according to Singwi[4] et al. to include the effect of exchange and correlation between conduction electrons to obtain the screened form factor. The model potential contains two arbitrary parameters which have been evaluated by fitting to experimental phonon frequencies at zone boundaries. Thus the calculation of form factor, energy wave number characteristic and Cochran's G(q) function is straight-forward and has been done for any arbitrary positions of ions.

The third chapter mainly deals with the computation of phonon dispersion relations in lithium, sodium, potassium and rubidium using the author's model potential and self consistent dielectric function of Singwi et al. The theoretical results have been found to be in good agreement with the experiment for all the metals taken for investigation. It is worthwhile to mention that the model potential also reproduces the observed crossing of transverse branch over the longitudinal one in (100) direction in case of lithium.

The chapter IV deals with the evaluation of interionic potential, interatomic force constants for various orders of neighbours and elastic constants using the model potential described in chapter II. The detailed behaviour of interionic potential has been discussed. It has been found that theminimum of interionic potential in all the metals lies in the region between first and second neighbours as neutron scattering data indicate. The calculated interatomic force constants upto fifth shell of neighbours have been found to be in reasonably good agreement with those obtained from the phonon data by a Born-von-Karman analysis. Using these force constants the elastic constants have also been evaluated and found to be in reasonable agreement with the experimental ones.

In Chapter V the author's model potential has been employed to calculate the resistivity and thermoelectric power of liquid metals (Li, Na, K and Rb). In computation of these properties the structure factors of the liquid metal play an important role. In this chapter the author has considered this point in detail. Experimentally known structure factors as well as theoretically derived from Percus-Yevik theory[7,8] have been used in the calculation and their effect on the calculated properties has also been studied. Recently highly accurate X-ray diffraction measurements for the static structure factors for liquid sodium (at 100° and 200° C) and liquid potassium (at 65° and 135°C) have been reported by Greenfield Wellendorf and Wiser[9]. The use of these structure factors in calculating the above properties shows an improvement over the results obtained using other structure factors. In the end it has been concluded that the accuracy of the model potential can be tested by calculating these properties only when the accurate measurements on the static structures are available.

The Chapter VI deals with the calculation of binding energy and paramagnetic susceptibility. The calculated values of these have also been found to be in reasonable agreement.

The Chapter VII deals with the theoretical study of lattice dynamics and some thermal properties e.g. specific heats, Debye temperatures and Debye Waller factors of gold based on a theoretical model. The theoretical results from this model are in better agreement than the previous theoretical studies. Finally this study emphasizes the need for a consideration of repulsive interaction for such class of metals.

It is evident from the present investigation that the proposed model potential successfully interprets almost all the properties of alkali metals and establishes in its own right its importance in the field of lattice dynamics. From the present study one may gain confidence in choosing this form of model potential and applying it to study other properties of simple metals.

A part of the above work has resulted in the following papers:-

1. Dispersion relations in gold; Phys.Stat.Sol. 43b, K153 (1971).
2. Lattice Dynamics of gold; Ind.J.Pure Appl. Phys. 10, 105 (1972).
3. Debye-Waller factor of gold; Sol.Stat.Comm. 13, 77 (1973).
4. A new model potential to study lattice dynamics of alkali metals (to be communicated to Physica Status Solidi 1974).
5. Effective ion-ion potential in alkali metals (to be communicated to Physica 1974).
6. Effect of Structure factor on electrical properties of sodium and potassium (to be communicated to J.Phys.F. 1974).

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