NOTIZEN

Modified Shell Model Treatment of Phonons in Metals

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Dedicated to Professor Dr. H. Maier-Leibnitz
on his 60th birthday

The lattice dynamics of solids can be considered from two different points of view. Formally, dispersion curves may be described by a set of force constants which reproduce the experimental results of neutron spectroscopy [1-6]. On the other hand, one can try to derive the dispersion relations from the electronic states in a microscopic theory [7-17].

It has turned out that a useful intermediate description is given by model theories which simulate the microscopic interaction by certain model parameters. For example, the shell model [18, 19, 20] and its modifications give a natural parametrization of the lattice forces in insulating crystals. Especially, the breathing-shell model [21, 22] was successful in deriving the dispersion curves of ionic crystals from macroscopic data. The main success of this model is to replace formal long-range ion-ion interactions by long-range dipole forces and short-range adiabatic electron-ion forces. This reduces the number of necessary parameters drastically.

Recently, it has been shown that a modified version of the breathing-shell model which is an extension of Cochran's [20] model can be applied successfully to homopolar substances with a high dielectric constant like Si, Ge and gray-tin [23]. From this, it arises the question whether the general features of such a model are valid for other crystals with an adiabatic electron-ion interaction, e.g. for metals. This would be especially useful for crystals like Pd, Ni, Cu etc. where a reasonable force-constant description of the dispersion curves needs at least interactions with third nearest neighbours. To be within the limits of experimental error fits have to be extended up to seventh and even higher order neighbours [3-6]. So far as microscopic (Cu [24]) or semi-phenomenological (Cu [25, 26]) calculations are available, there are still discrepancies of the order of $10-20\%$. It is clear from the theoretical treatments that the formal force constant description simulates the microscopic mechanism in a complicated way. In this respect the situation is reminiscent to that in insulating crystals like Ge [23, 27, 28].

We have tried to apply the idea of an adiabatic shell model to the lattice dynamics of metals. From the experience in insulating crystals we used only a monopol-(breathing) and a dipole-mode for the adiabatic response of the electronic system. Fig. 1 shows the model force constants for a face-centered metal. The notation follows that used in ionic crystals [22].

In our calculation three parameters are fitted to the elastic constants. The other four parameters are obtained from the experimental points at LA(X), TA(X), LA(L) and TA(L). The results for Ni are shown in Fig. 2. Similar results are obtained for Pd and Cu and Al. The parameters used are given in the table:
There are interesting features in the results. For all the metals discussed the used ionic charge is near to one. That means that only a very small part of the

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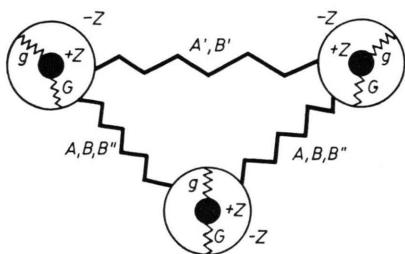[28] B. Szigeti, to be published in Proc. Phys. Soc. London.

Fig. 1. Force constants of a breathing shell model for a face-centered cubic crystal. A, B nearest neighbour force constants, B'' non-central part of B, A', B' second nearest neighbour force constants, Z core charge ($= $ -shell charge), G shell-core dipole spring, g shell-core breathing spring.

	A	B	A'	B''	G	g	Z
Ni	42.37	2.68	1.19	−5.11	128.0	330.33	1.115
Cu	32.0	2.725	0.575	−4.725	86.043	200.067	1.197
Pd	42.5	5.14	1.445	−7.64	108.38	231.858	1.144
Al	13.57	0.51	2.94	−2.52	270.0	109.90	1.0
Na	1.865	0.004	0.256	−0.051	100.0	421.54	0.5

outer electrons is taking part in the adiabatic polarisation screening. Most of the electrons behave as if they are coupled rigidly to the ions. This corresponds to the result obtained by PRAKASH and JOSHI [14] for alu-

minum that a microscopic treatment gives better results if only one electron is treated in a dynamical way while the other two valence electrons are considered to be bound to the ionic core. All the parameters used in our model are nearest neighbour springs besides of A' which turns out to be very small. We checked the applicability of the model to bcc metals. In the special case of sodium we obtained a very good fit with the parameters given in the table. The ratio of A/G is very small (10^{-2}) compared with values of about $1/3$ for the transition metals of the table. This indicates that in metals with nearly free electrons like Na — and, to a smaller extent Al — the dipole forces and the polarisation effects are less important compared with the rigid ion part. The results of this treatment supports the conjecture that for many metals a phonon-theory can be constructed which considers them as built from pseudo-atoms with rather tightly bound electrons. As long as the Kohn anomaly is unimportant for the phonons in a metal there is no basic argument against such a point of view. We think that our investigation might be a helpful link to the right form of a microscopic theory even for complicated crystals like transition metals.

NICKEL 296 °K

●●● exp. dispersion curves (Birgeneau et. al.)
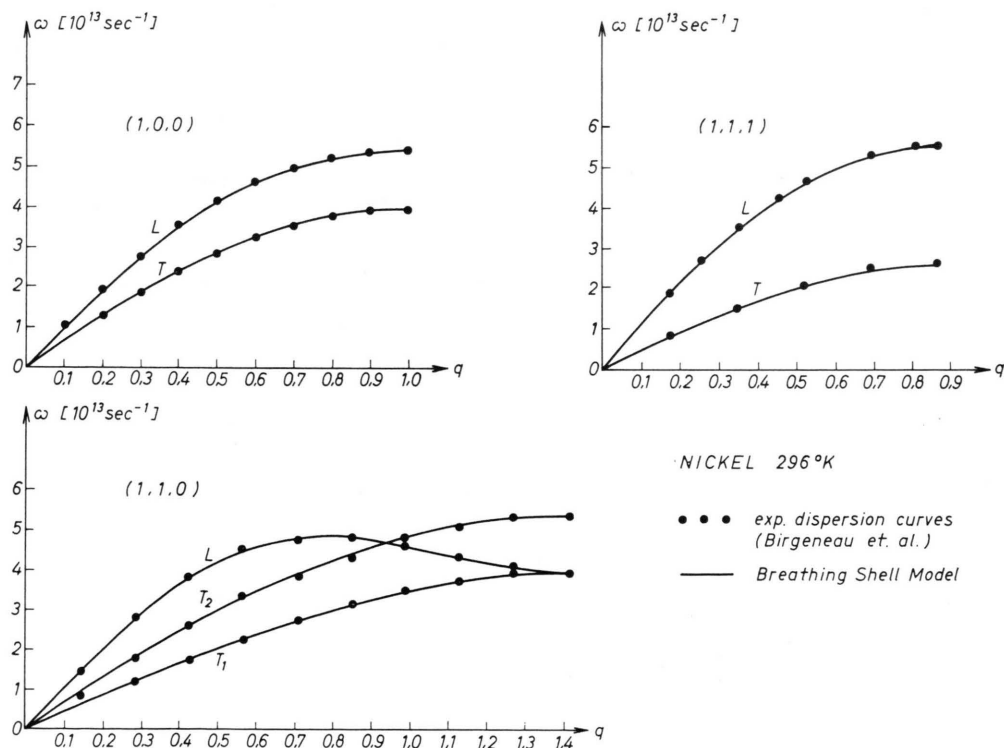
——— Breathing Shell Model

Fig. 2. Dispersion curves of fcc nickel at 296 °K. Parameters $A = 42.37$, $A' = 1.19$, $B = 2.63$, $B'' = -5.11$, $Z = 1.115$, $G = 128.0$, $g = 330.33$.