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Damping of Phonons in Simple Metals

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Calculations of the phonon damping due to the electron-phonon coupling are made in ten metals—Li, Na, K, Rb, Cs, Al, Pb, Be, Mg, and Zn—so as to cover all important structures, using Heine–Abarenkov–Animalu pseudopotential and different dielectric screening schemes. Screening is done in the spirit of the local reduction of the pseudopotential with proper considerations for the conduction-electron effective mass m^*. Comparison with experiments is made for those metals for which experimental results are available and it is found that a substantial contribution to the phonon damping arises from electron-phonon interaction. Li and Cs have a strange behavior in the $(\zeta,0,0)$ direction, where the damping becomes enormous for certain values of q in the small-q region. From an analysis of the results it is legitimate to divide the different screening theories into two groups. Random-phase-approximation (RPA) theory, Hubbard's modified RPA, and the Geldart–Vosko modification of Hubbard's theory are in the first group. All of these theories have weak screening effects. The second group contains the theories presented by Singwi *et al.*, Kleinman and Langreth, and Toigo and Woodruff. All of these theories have strong screening effects. Overhauser's theory has an intermediate screening effect.

I. INTRODUCTION

In recent years the pseudopotential approach has found increasing applicability in the theory of metals[1] and attempts have been made to study the phonon dispersion in a variety of metals.[2–10] However, relatively fewer attempts are made towards the measurement of the lifetime of individual phonons.[6,8,9] Since in metals the electron-phonon interaction plays an important role and causes appreciable forces among distant atoms, study of the phonon lifetime against the decay due to electron-phonon interaction offers a problem of inherent interest.

Experiments have shown that the damping is more for the longitudinal phonons than for transverse, particularly at low temperatures, where, as pointed out by Nilson and Stedman,[8] the major contribution to the phonon damping arises from electron-phonon interaction and to a lesser extent from anharmonic effects. Although qualitative calculations of the phonon damping due to electron-phonon interaction and anharmonicity have been made by a number of authors,[6,11–15] quantitative calculations are performed only for Al[16] and K.[5,6] Since then, there have also been many developments in pseudopotential[3,14,17] and dielectric screening theories.[18–22] It therefore seems to be fruitful to present computations based on these recent theories.

The calculations are performed using the Heine–Abarenkov–Animalu (HAA) pseudopotential,[17] which has previously been found to give very good results for phonon dispersion.[1] Our aim has been to investigate the effect of the electron-gas screening function on the electron-phonon coupling and to study how a single theory can reveal largely differing coupling effects in different metals—varying from bcc to fcc and to hcp. Throughout the present calculations we work in the harmonic approximation and thus restrict ourselves to low temperatures only.

In Sec. II we give a brief account of the theoretical background needed for the calculations together with the pseudopotential and various forms of dielectric screening. Calculations and discussions of the results are described in Sec. III and Sec. IV contains a summary and conclusions.

II. THEORY

The forces between the ions in a metal arise from three distinct sources: (i) direct Coulomb repulsion between ions of charge $+Ze$ immersed in a uniform compensating background to give charge neutrality; (ii) exchange forces due to core-wave-function overlap; and (iii) ionic interaction via the conduction electrons which try to screen the ions. Hence the dynamical matrix D is the sum of three terms arising from these sources,

$$D = D^C + D^R + D^E .$$

The overlap part D^R gives a negligible contribution[2] for alkali and simple polyvalent metals, normally referred to as simple metals. We limit our calculations to these metals. Thus, the matrix D can, in terms of the effective interaction v_{eff}, be written

$$D(\vec{q}) = \frac{1}{M\Omega} \sum_{\vec{h}} \left[(\vec{q}+\vec{h})(\vec{q}+\vec{h})\, v_{\text{eff}}(\vec{q}+\vec{h}) - \vec{h}\vec{h}\, v_{\text{eff}}(\vec{h}) \right] , \tag{1}$$

where M is the ionic mass, Ω is the volume of the lattice cell, and the summation extends over all the reciprocal-lattice vectors $\vec{h}$. When discussing the phonon lifetime, one has to go beyond the adiabatic approximation and the matrix D becomes complex; the damping is related to the imaginary part of this matrix. Within this framework the full width due to electron-phonon coupling is given by[5]

$$2\Gamma(\vec{q}) = \frac{m^* \omega_p^2}{4\hbar k_F^2} \sum_{\vec{h}} \left(\frac{|\vec{e}_s \cdot (\vec{q}+\vec{h})|^2}{|\vec{q}+\vec{h}|^2} G(|\vec{q}+\vec{h}|) - \frac{|\vec{e}_s \cdot \vec{h}|^2}{h^2} G(h) \right), \quad (2)$$

where $\vec{e}_s$ is the phonon polarization vector of branch s and ω_p is the plasma frequency. $G(q)$ is expressible in terms of the pseudopotential $W_0(q)$ and the dielectric function $\epsilon(q)$.

The HAA model potential[17] is given by

$$V(r) = -\sum_l A_l(E) P_l \qquad (r < R_m)$$
$$= -Ze^2/r \qquad (r \geqslant R_m) \qquad (3)$$

plus some orthogonality and correlation corrections which are of local form. The different symbols have their usual meaning.[17]

The Fourier transform of this potential leads to the unscreened form factor consisting of the local part—$V(q)$ (K independent)—and a nonlocal part—$F(q, K, K')$ (K dependent),

$$W_0(K, q) = V(q) + F(q, K, K') . \qquad (4)$$

The local bare-ion potential is

$$V(q) = -\frac{4\pi}{\Omega q^2} \left[\frac{2A_2}{q} (\sin y - y \cos y) + 2Z \cos y + \left(\frac{|\mu_c(n_b)|}{q} - \frac{6Z\alpha_{\text{eff}}}{z^2} \right) (\sin z - z \cos z) \right] . \qquad (5)$$

Here $y = qR_m$, $z = qR_c$, Z is the valency, μ_c the chemical potential, α_{eff} the orthogonalization charge which equals $\frac{1}{2}(R_c/R_a)^3$, and the last term comes from the orthogonality and correlation corrections.

The nonlocal bare-ion potential $F(q, K, K')$ is evaluated by considering the scattering only from the initial states on the Fermi surface. Thus for $|\vec{q}| \leqslant 2k_F$, we set $K = K' = k_F$ and

$$F(q, K) = -\frac{4\pi R_m^3}{\Omega} \left[(A_0 - A_2) \left([j_0(a)]^2 - \frac{\cos a}{a} j_1(a) \right) + 3(A_1 - A_2) \frac{2k_F^2 - q^2}{2k_F^2} \{ [j_1(a)]^2 - j_0(a) j_2(a) \} \right] , \qquad (6)$$

where the j_l are spherical Bessel functions, $a = k_F R_m$, and $a' = K' R_m$.

For $|\vec{q}| > 2k_F$, we take $K = k_F$, $K' = q - k_F$, and

$$F(q, K) = -\frac{8\pi R_m^2}{\Omega [k_F^2 - (q - k_F)^2]} \{ (A_0 - A_2) \times [k_F j_1(a) j_0(a') - (q - k_F) j_1(a') j_0(a)] - 3(A_1 - A_2) [k_F j_2(a) j_1(a') - (q - k_F) j_2(a') j_1(a)] \} . \qquad (7)$$

The screening of $W_0(K, q)$ is done in the spirit of the local reduction of the potential inspired by the results of Pindor and Pynn,[23] who have obtained qualitatively good results for the lattice dynamics of Mg from this approximation.

Further, first-principle calculations of the phonon damping require a proper treatment of the conduction-electron screening function, since once $\epsilon(q)$ is known we may directly use Eq. (2) to determine the phonon damping. We now elucidate different forms of the dielectric screening used in our calculations and the relative merits and drawbacks of each.

A. Random-Phase-Approximation (RPA) Theory

In this approximation[24] the response of the electrons to the influence of the Hartree potential is of a free-particle type and effects due to exchange and correlations are neglected. Since the orthogonality correction in Eq. (5) introduces an extra charge of the order of $Z\alpha$ around each atomic site, a correction for this has to be made in the dielectric function itself. With this correction, the dielectric function in the RPA becomes

$$\epsilon(q) = 1 + (1 + \alpha_{\text{eff}}) Q_0(q) , \qquad (8)$$

where

$$Q_0(q) = \frac{2k_F}{\pi a_0 q^2} \left(1 + \frac{4k_F^2 - q^2}{4k_F q} \ln \left| \frac{q + 2k_F}{q - 2k_F} \right| \right) \qquad (9)$$

is the usual Lindhard function. In this treatment no considerations are made for the effects of the lattice on conduction electrons. This may roughly be done by introducing an effective mass m^*; for an exact treatment of this, however, one has to appeal to higher-order perturbation theory which is beyond the scope of this paper. The dielectric function then becomes

$$\epsilon(q) = [1 + \mu S_0(q)] , \qquad (10)$$

with

$$S_0(q) = Q_0(q)(1 + \alpha_{\text{eff}})$$

and

$$\mu = m^*/m . \qquad (11)$$

B. Modified Hubbard (HB) Theory

Hubbard[25] has modified the expression for the dielectric function by taking into account the effects of exchange between electrons whose spins are parallel. The expression then reads

$$\epsilon(q) = 1 + (1 + \alpha_{\text{eff}}) \mu Q(q) , \qquad (12)$$

with

$$Q(q) = Q_0(q)/[1 - f(q)\,Q_0(q)]\ . \qquad (13)$$

The factor $f(q)$, which is really the crux of a screening theory, is given in this case by

$$f(q) = \tfrac{1}{2}\,[q^2/(q^2 + k_F^2 + k_{\mathrm{TF}}^2)]\ , \qquad (14)$$

where k_F is the Fermi radius and k_{TF} is the Thomas–Fermi screening factor,

$$k_F = (3\pi^2 Z/\Omega)^{1/3} \quad \text{and} \quad k_{\mathrm{TF}}^2 = (4k_F/\pi a_0)\,\mu\ .$$

C. Geldart and Vosko (GV) Theory

Although the HB dielectric function adequately includes exchange effects, this form, together with the RPA, does not satisfy the compressibility sum rule because of the exclusion of short-range Coulomb correlations. GV modified the Hubbard expression by introducing a parameter ξ so as to get the correct $q = 0$ limit of the dielectric function. Equation (14) is replaced by

$$f(q) = \tfrac{1}{2}\,[q^2/(q^2 + \xi\, k_F^2)]\ . \qquad (15)$$

In the interpolation scheme of Nozieres and Pines, ξ is given by

$$\xi = 1/(0.5 + 0.013\mu r_s)\ . \qquad (16)$$

Here r_s is the interelectronic distance, a property of the individual metal computed from

$$r_s = (3\Omega/4\pi Z)^{1/3}\ .$$

One finds that this $f(q)$ is inadequate in the region $q \sim k_F$, but a possible way of surmounting this shortcoming has been put forward by Geldart and Taylor.[22]

D. Singwi, Sjolander, Tosi, and Land (SSTL) Theory

While the dielectric function of GV is physically reasonable in that it includes exchange effects, it has the unsatisfactory feature that for large values of q, i.e., small values of r, the pair correlation function becomes negative, a completely unphysical result. This has recently been corrected for by Singwi *et al.* in a series of papers,[18] hereafter referred to as SSTL I and II. They have given a self-consistent procedure for calculating the dielectric function through the decoupling of the two-particle distribution function. These theories give remarkable improvement over the earlier ones with respect to the behavior of different physical quantities. A comparison of the two reveals that SSTL I gives better results for $g(r)$ than SSTL II but only at the cost of sacrificing the compressibility sum rule. An interpolation has been attempted between the large-q behavior of the former and the small-q behavior of the latter, in the form

$$f(q) = A(1 - e^{-B(q/k_F)^2})\ , \qquad (17)$$

which gives a good approximation to the self-consistent $f(q)$ and deviates from the latter only for $q > 2k_F$, where $\epsilon(q)$ itself becomes less sensitive to the exact form of $f(q)$ because of the smallness of $Q_0(q)$.

E. Kleinman and Langreth (KL) Theory

The problem of the dielectric screening has been treated by Kleinman[19] through a diagrammatic technique and later by Langreth[19] via a variational approach taking into account the Hermitian property of various types of interactions. They obtained an expression for the dielectric function and the vertex function in a closed form. In the static limit the two expressions, however, have the same form,

$$f(q) = \frac{1}{4}\left(\frac{q^2}{q^2 + k_F^2 + k_s^2} + \frac{q^2}{k_F^2 + k_s^2}\right)\ , \qquad (18)$$

where k_s is the screening parameter expressible in the Nozières and Pines interpolation scheme by

$$\frac{k_s^2}{k_F^2} = \frac{1 - 0.0395(k_{\mathrm{TF}}/k_F)^2}{1 + 0.0395(k_{\mathrm{TF}}/k_F)^2}\ . \qquad (19)$$

The KL scheme also satisfies the compressibility sum rule properly, but gives negative values of $g(r)$ over a considerable range of the interparticle separation.

F. Overhauser (OV) Theory

OV[20] has tackled the problem of the electron-gas polarizability from a different standpoint. The essence of his theory is the replacement of the complete excitation spectrum of the electron gas by a single plasmon branch $\hbar\omega(q)$; for small q, $\omega(q)$ approaches the plasma frequency, and for large q it approaches the free-electron limit $\hbar q^2/2m$. This model gives realistic behavior primarily in the intermediate-q region where the spectral width of the one-electron excitation is comparable to $\hbar\omega(q)$. The model at the outset is chosen so as to satisfy several important physical requirements. He chooses $f(q)$ to be of the form

$$f(q) = 1.1x^2/(1 + 10x^2 + 1.5x^4)^{1/2}\ , \qquad (20)$$

where $x = q/2k_F$.

This $f(q)$ satisfies the limiting condition of the pair correlation function $[g(0) \sim 0.1$ and hence $f(\infty) \sim 0.9]$ and gives values of the correlation energy comparable to the published results.[18] The form is actually SSTL-II-model dependent and a phenomenological one because the coefficient, 10, of the x^2 term in Eq. (20) is derived from a comparison of the correlation energy to that of SSTL II at $r_s = 4$. However, a striking difference in the two is that this $f(q)$ does not rise to its limiting value as sharply as that of SSTL II (and also of GV). SSTL have shown that a fast rise in $f(q)$ vs q prevents $g(0)$ from being negative, in spite of the fact that their $f(q)$ does not give the correct limiting behavior.

G. Toigo and Woodruff (TW) Theory

A more recent attempt in this direction has been made by TW,[21] who give a general expression for $\epsilon(q, \omega)$. In the static limit ($\omega = 0$) their expression is also a functional of $f(q)$ for which they derive an expression from first principles with no adjustable parameters. This $f(q)$ properly satisfies the compressibility sum rule and the asymptotic limit

$$\tfrac{1}{2} \leq f(\infty) \leq 1 \; .$$

Their calculation is based on a straightforward method for decoupling and solving the equation of motion for the time-transformed Green's function for the charge-density operator. The frequency moments of all the spectral functions involved are also conserved. Here

$$f(q) = P_0(q)/S_0(q) \; , \tag{21}$$

where $P_0(q)$ includes exchange and correlation effects and $S_0(q)$ is the same as defined in Eq. (11). This $f(q)$ is more reliable for small and large values of q and, above all, is correct in exhibiting a peak for $x < 1$ which SSTL and HB do not. The function $P_0(q)$ is determined through integrals whose calculation is very cumbersome. We have, therefore, generated the values of $f(q)$ at the required q values via a quadratic interpolation from the tabulated values of these authors.

Having thus defined the pseudopotential and the dielectric screening function, the function $G(q)$ appearing in Eq. (2) is expressible as

$$G(q) = \frac{1}{2x}\left(\frac{k_{\mathrm{TF}}\, W_0(q)}{qQ_0(q)\, W_c(q)} \; \frac{\epsilon(q) - 1}{\epsilon(q)}\right) \; , \tag{22}$$

where $W_c(q)$ is the Fourier-transformed Coulomb potential. We now present the calculations of phonon damping based on the above theories.

III. RESULTS AND DISCUSSIONS

Using the expressions given in Sec. II for the pseudopotential and various dielectric screening $f(q)$'s, numerical calculations are performed for the phonon full width at half-height due to the electron-phonon interaction in ten metals. The metals chosen are alkali metals with bcc structure; Al and Pb with fcc structure; and Be, Mg, and Zn with hcp structure, so as to cover the most commonly occurring and important types of structures. The values of different physical constants for each metal used in these calculations are listed in Table I. Numerical computations were performed on the IBM 360/44 computer at Delhi University, Delhi.

In order to make the discussions of the results obtained in this paper tractable we have classified the metals into three groups according to the type of the structure, particularly because it would not be easy to discuss the three groups on the same footing. As well known, the electron interaction with the ions and phonons is drastically different in these three classes. Nevertheless, before turning our attention to the individual classes, it is worthwhile to make some general remarks about the over-all character of the phonon damping and justify the approximations used in the calculations.

Many computations of the phonon dispersion in metals have been reported[6,17] using the local HAA pseudopotential and also with the modified versions,[4,23] which take into account nonlocal effects and corrections for short-range nonpair forces. In all these calculations only slight improvements in the results could be obtained, and those too at the cost of introducing a number of adjustable parameters. Moreover, most of these results could be reproduced quite simply by using local potentials alone. On the basis of this we anticipate that HAA potential, as discussed in Sec. II, may give a fair description of the situation provided proper considerations are made for exchange and correlation effects. While the Ashcroft pseudopotential[26] itself gives fair results of the lattice dynamics, we expect that from the HAA potential, which is inherently superior[27] because of the absence of large discontinuities at $r = r_c$ (core radius), the results would be even more reliable. It is therefore worth emphasizing that for the discrepancies encountered in the computations one has to seek explanation other than only the exclusion of nonlocal effects. One possible factor that may affect the results may be the presence of the many-body effects in electron correlations at high densities. To some extent these effects have been included in the more recent theories (SSTL, KL, OV, and TW).

Even more important appears to be the influence of the effective mass m^* of the conduction electron in order to include the band effects, which we have taken into account in an over-all manner, as discussed earlier. Nevertheless, as suggested by Weaire,[28] it might be possible to include the effects of some of the higher-order terms of the perturbation series by adjusting m^*. Further, the use of m^* implies taking an effective k_F^* for the range of integration over the Fermi surface to obtain the appropriate screening field. This partly accounts for the nonsphericity of the Fermi surface.

A serious limitation of the present endeavor arises because of the nonavailability of any direct experimental results on the phonon damping corresponding to the electron-phonon interaction, which to some extent limits the achievement of our objective. The basic difficulty in measuring it from neutron-scattering experiments is the contribution from anharmonic effects[6] which are sig-

TABLE I. Values of different parameters used in the calculations.

	LI	Na	K	Rb	Cs	Al	Pb	Be	Mg	Zn
a^{a}(Å)	3.46	4.24	5.333	5.62	6.05	4.042	4.941	2.283	2.268	2.659
c^{a}(Å)	⋯	⋯	⋯	⋯	⋯	⋯	⋯	3.607	3.594	4.934
k_F	0.590	0.488	0.395	0.370	0.341	0.928	0.835	1.029	0.7242	0.834
m^*	1.19	1.01	0.99	0.197	0.98	0.94	0.91	1.28	1.01	0.93
r_s	3.258	3.131	4.862	5.197	5.625	2.069	2.298	1.866	2.850	2.301
R_0	1.47	1.85	2.51	2.82	3.12	1.08	1.59	0.64	1.47	1.57
R_m	2.8	3.4	4.2	4.4	4.8	2.0	2.1	2.0	2.6	2.2
α_{eff}	0.046	0.052	0.069	0.080	0.085	0.024	0.041	0.010	0.043	0.079
$-\mu_c$	0.0914	0.0837	0.0734	0.0700	0.0658	0.0103	0.0101	0.1053	0.0975	0.1010
A_0	0.336	0.305	0.240	0.224	0.205	1.380	1.920	1.010	0.780	0.990
A_1	0.504	0.339	0.256	0.226	0.207	1.640	2.000	1.220	0.880	1.140
A_2	0.455	0.402	0.368	0.384	0.366	1.920	0.900	1.480	0.990	0.980
ξ	1.84	1.81	1.79	1.79	1.78	1.90	1.90	1.88	1.87	1.89
A^{b}	0.999	0.995	1.007	1.008	1.011	0.907	0.925	0.888	0.947	0.925
B^{b}	0.258	0.263	0.249	0.247	0.242	0.336	0.324	0.349	0.307	0.324
$\nu_p{}^{c}$	17.60	5.58	4.07	2.49	1.77	29.89	10.61	52.30	20.30	14.30

[a] *Handbook of Chemistry and Physics* (Chemical Rubber Publishing Co., Cleveland, Ohio, 1959).

[b] Reference 18.

[c] In units of 10^{+12} Hz calculated from $\nu_p^2 = Z^2\, e^2/\pi M \Omega$. All other parameters are taken from Ref. 17 and are in a.u.

nificant even at 0 °K due to the zero-point motion. Further restrictions in the absolute measurement of the width arise from the limitations of the instrumental resolution. Nevertheless, inferences may be obtained from the comparison of the calculated results with the observed phonon width and the phonon frequencies.

A comparison of the various versions of the free-electron dielectric screening has been made in Fig. 1 where we have plotted the dimensionless quantity $1/[1-f(q)\,Q_0(q)]$ vs q for Na. The figure shows that the various dielectric response functions differ substantially for small values of $|\vec{q}|$ and tend to match at higher values of $|\vec{q}|$. This indicates that the choice of the screening function influences the results where small-momentum-transfer processes dominate over large-momentum-transfer processes.

We now discuss each group of metals separately and emphasize the relevant points. A judicious selection is made in presenting the results because of similar behavior in a number of cases; results are plotted only for those metals (a) for which the experimental results are available and (b) which show a peculiar behavior. However, the discussions will be extended to other metals also, when it seems meaningful.

A. bcc Metals

The first set of metals chosen for these calculations is the alkali metals. The importance of these metals lies in the fact that their structure is simple from a theoretical point of view[5] and there is a large amount of experimental data as well.[6,7] This group forms a very nearly-free-electron system and the core-repulsion interaction is too small since the ion radii[29] are considerably smaller than the interionic distances. Thus the basic assumptions mentioned in the theory are very nearly satisfied. Summation over the reciprocal-lattice vectors is limited to those points for which $|\vec{h}| \leq 7k_F$ and the calculations are performed for three

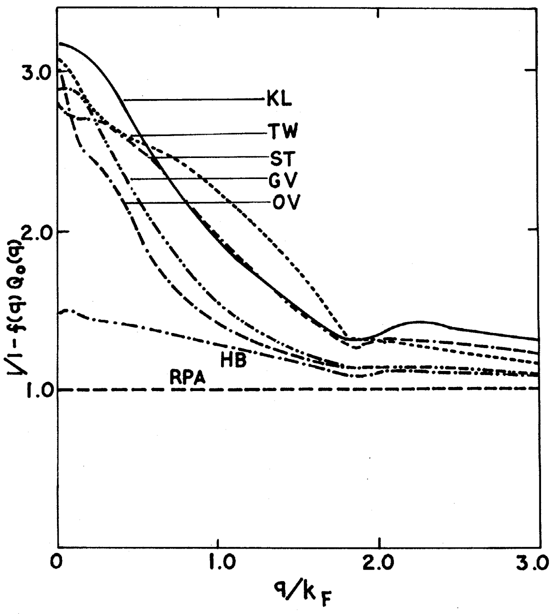

FIG. 1. Dimensionless quantity $1/[1-f(q)Q_0(q)]$ from different dielectric-screening theories for Na. Notations used are RPA, random-phase approximation; HB, modified Hubbard theory; GV, Geldart–Vosko theory; ST, Singwi–Sjolander–Tosi–Land theory; KL, Kleinman–Langreth theory; OV, Overhauser theory; TW, Toigo–Woodruff theory. The same notations are used everywhere.

high-symmetry directions—(ζ, 0, 0), (ζ, ζ, 0), and (ζ, ζ, ζ)—for both the longitudinal and transverse branches. We have not shown the results of the T_1 branch in the (ζ, ζ, 0) direction and both the T_1 and T_2 branches in the (ζ, ζ, ζ) direction since the values of Γ come out to be extremely small for these polarizations (10^{-2}–10^{-3} times smaller than the others).

Results for the alkali metals are shown in Figs. 2–4 for the (ζ, 0, 0) direction in Li and Na [also for the (ζ, ζ, ζ) direction] and for all the directions in K. In the case of Li we find a pronounced damping in the (ζ, 0, 0) direction for both the longitudinal and transverse branches near $q = 0.3$ (in units of $2\pi/a$, where a is the lattice constant). A situation akin to this also arises in Cs and a little less strongly in Rb at $q = 0.2$. In contrast, no such behavior is observed in Na and K. A possible reason for this might be a greater role of the band effects in Li, Rb, and Cs. Thus the second-order-perturbation corrections to the band gaps are significant in Rb and Cs as there are d states near the Fermi level. It is also known[5] that the inclusion of the nonlocal part in the pseudopotential is more important for Li, Rb, and Cs than for Na and K. A typical behavior has also been shown by Li in the phonon-dispersion curves and the attempt to construct an effective pseudopotential to explain this has so far failed.[7] Besides, similar differences occur in treating these metals as completely-free-electron systems. Theoretical and experimental results for energy bands[30] reveal that the Fermi surface deviates from sphericity only in Li, Rb, and Cs. Further, in Cs the relativistic effects may start showing up and the larger ionic radii of Rb and Cs may cause exchange-overlap contributions to be appreciable. Owing to the lack of any experimental results on these metals, no assessment can be made on the consistency of the results.

Our results for Na and K are comparable to those published previously,[5,6,13] having maxima at about $q = 0.4$ in the (ζ, 0, 0) L branch. We can compare

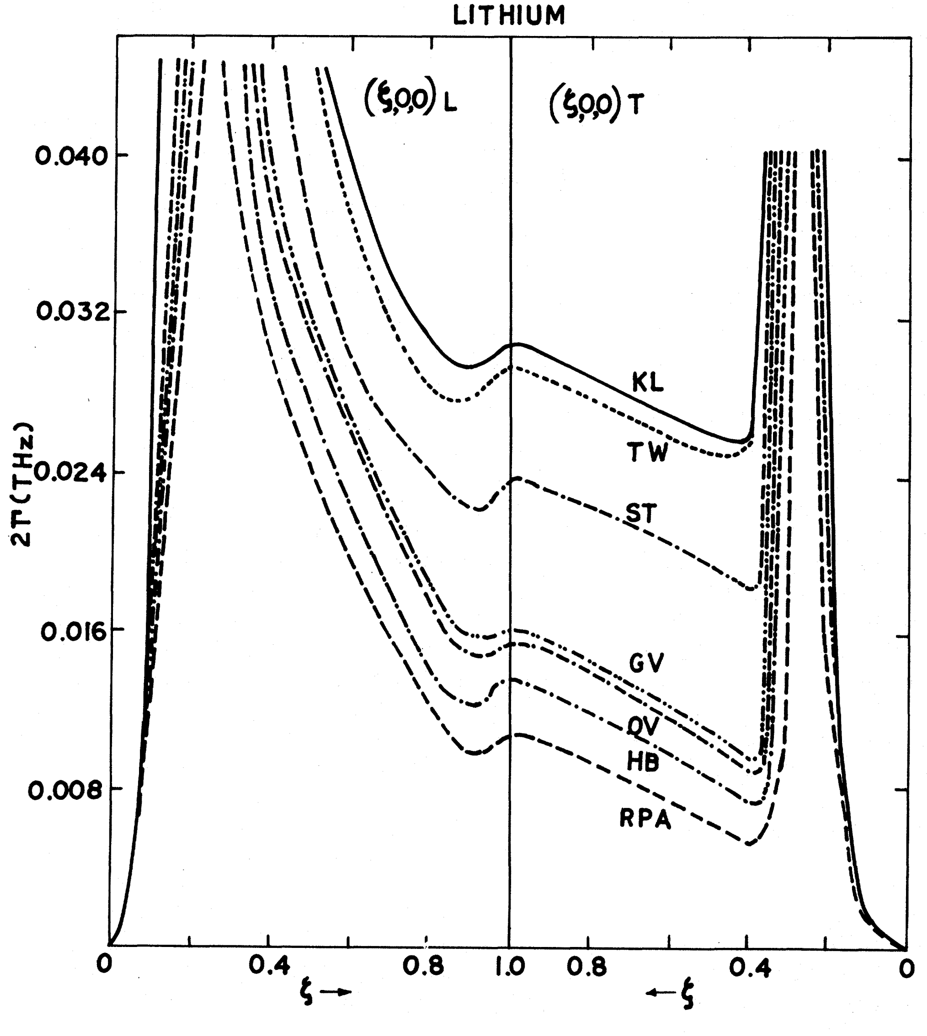

FIG. 2. Phonon full widths at half-height due to electron-phonon coupling vs phonon wave vector ζ in Li for (ζ, 0, 0) direction.

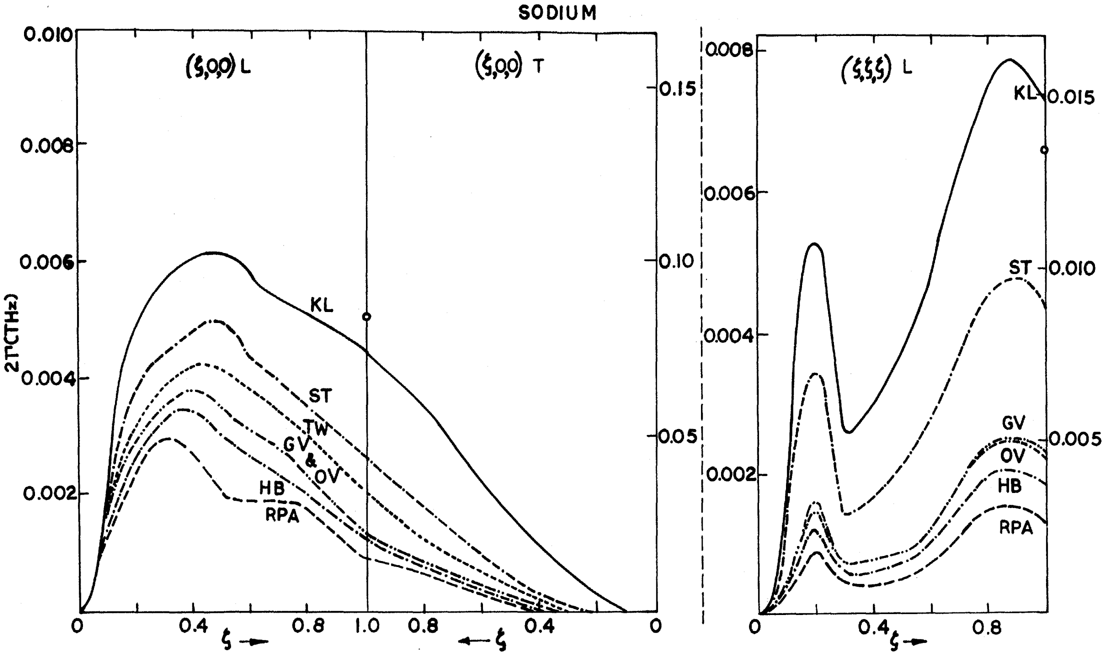

FIG. 3. Same as in Fig. 2 for Na for $(\zeta, 0, 0)$ and (ζ, ζ, ζ) directions, ○ are the experimental points shown on the right-hand side scale. The experimental data are sampled from Ref. 13.

these results with the experimental results which have been reported for these two cases.[6,13] Particularly, it should be kept in mind that while we are considering only the electron-phonon coupling, the experimental results also incorporate the anharmonicities so that agreement may be expected to an order of magnitude. In fact, our calculated values are about half the experimental ones. The damping is much stronger in the $(\zeta, \zeta, 0)$ direction as compared to both the $(\zeta, 0, 0)$ and (ζ, ζ, ζ) directions, but the difference becomes smaller as we go to the higher member of the series. Li shows a startling feature in that the strength of the coupling is higher by almost an order of magnitude over the others, a reflection of the strength of the pseudopotential. This is also exhibited in the dispersion curves.[5] In all cases, the width of the T_2 branch increases steeply at about $q = 0.38$, a manifestation of the Kohn anomaly in the phonon lifetime. It may be noted that vast differences in the T_1 and T_2 branches of the $(\zeta, \zeta, 0)$ direction have been observed in the disperion curves too.[2] A situation opposite to this arises in bcc transition metals[31] (Nb, Mo, Ta, and W) where the two transverse branches of the $(\zeta, \zeta, 0)$ direction almost overlap.

B. fcc Metals

The next group of metals chosen for our study is Al and Pb possessing a fcc structure. This choice is dictated by the availability of the experimental results of phonon damping.[9,16] Another interesting feature is that in spite of having the same structure, the strength of the electron-ion coupling is drastically different in the two metals and also different from that in alkali metals. As well known, here the core size is also very small and the conduction-electron states are plane-wave-like over most of the volume. Summation in the reciprocal space is restricted to the points for which $|\vec{h}| \leq 6k_F$ and the calculations are carried out for the same high-symmetry directions as in bcc metals, i.e., $(\zeta, 0, 0)$, $(\zeta, \zeta, 0)$, and (ζ, ζ, ζ).

Result for Al and Pb are given in Figs. 5 and 6, respectively. For the $(\zeta, 0, 0)$ L direction in Al the damping rises abruptly at $q = 0.7$ and then decreases monotonically, whereas in the transverse branch it is almost zero except in the narrow range of $q = 0.6$–0.8 with a peak at $q \sim 0.7$. The major contribution to the longitudinal branch is expected from the $h = 0$ term (the normal process). This phenomenon is seen to be much more marked in Al than in all other cases, in that almost half the contribution comes from this term. In contrast, in Pb the $h \neq 0$ term (the umklapp process) is more significant so much so that the T_2 branch of the $(\zeta, \zeta, 0)$ direction goes over the longitudinal branch. In both cases for the $(\zeta, \zeta, 0)$ T_1 branch we encounter strong peaks for certain values of q but otherwise the damping is negligible. This gives strong evidence of the Kohn anomaly. Along with the fact that a number of Kohn anomalies are

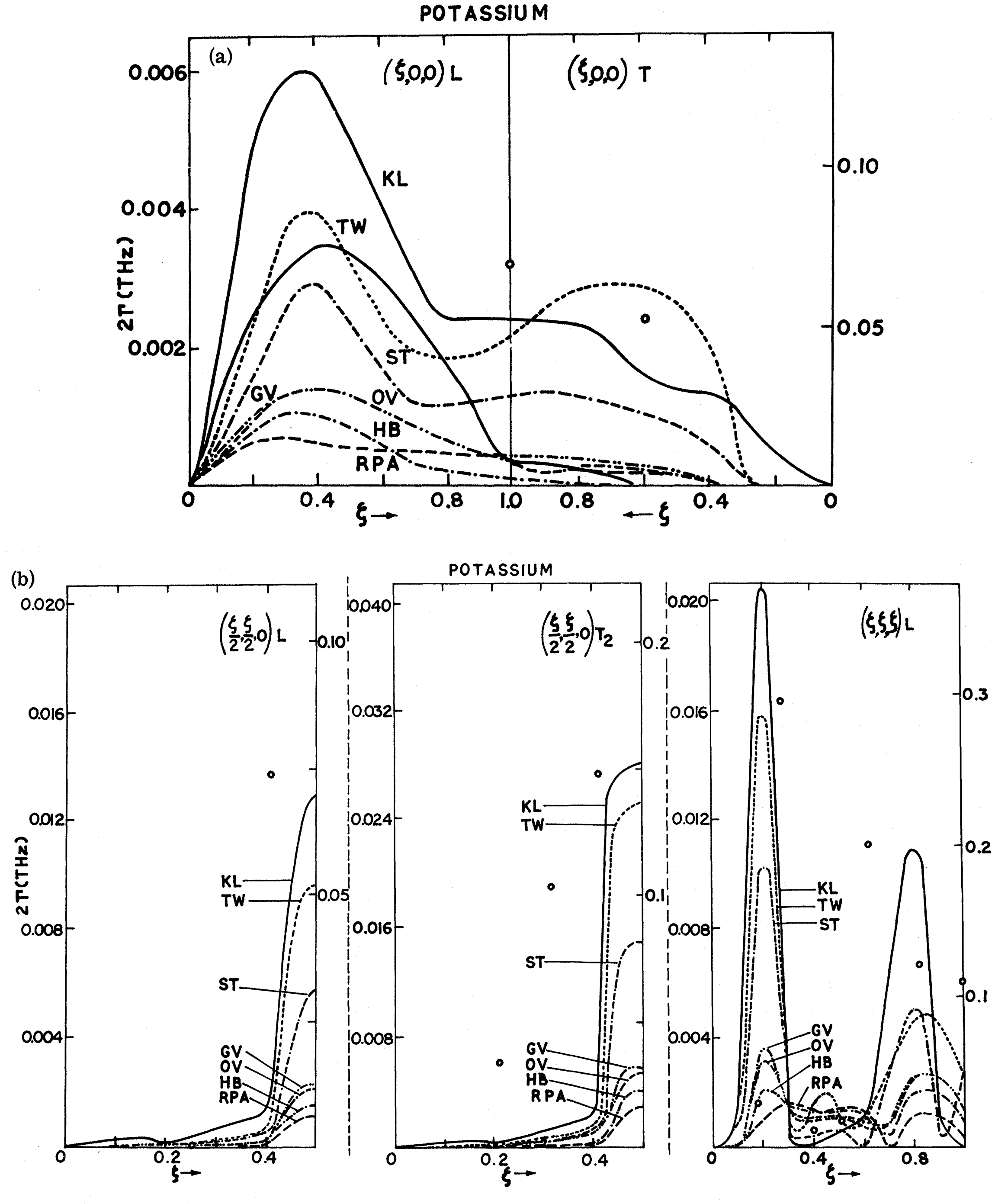

FIG. 4. (a) Same as in Fig. 3 for K in (ξ, 0, 0) direction. The experimental data are sampled from Ref. 6. (b) Same as in (a) for (ξ, ξ, 0) and (ξ, ξ, ξ) directions.

clearly visible in Pb, there is a considerable difference in the over-all damping behavior of the two; damping is larger in Pb than in Al by almost an order of magnitude indicating that the strength of the electron-ion coupling is enormous in Pb as compared to Al. In particular, many dips are seen in the (ξ, 0, 0) and (ξ, ξ, 0) L branches in Pb; the source may possibly be the presence of strong long-range oscillatory forces. Even more striking is the difference in the electron-screening behavior because of the higher valence of Pb where the electrons screen the ion motion more effectively.

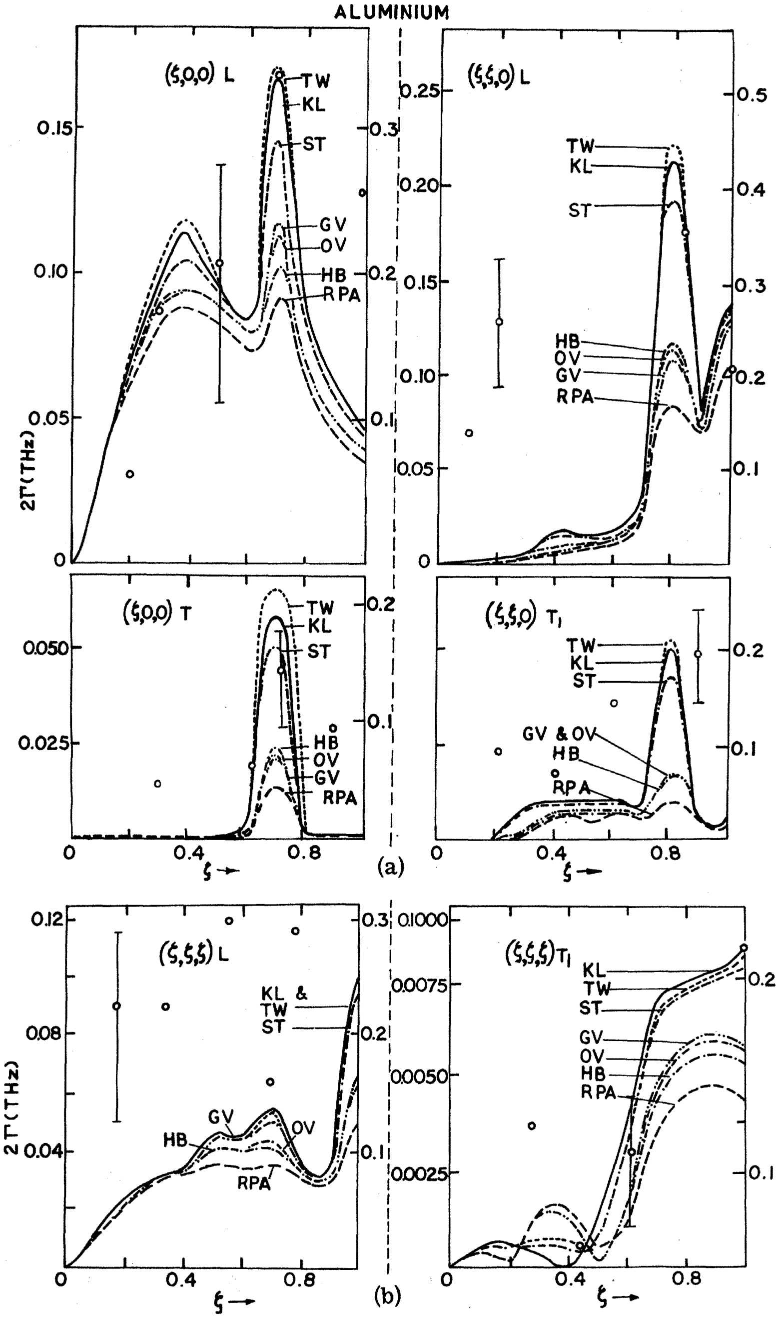

FIG. 5. (a) Same as in Fig. 3 for Al in $(\zeta, 0, 0)$ and $(\zeta, \zeta, 0)$ directions. The experimental data are sampled from Ref. 16. (b) Same as in (a) for (ζ, ζ, ζ) direction.

C. hcp Metals

The phonon-lifetime calculations for the hcp structure are made for Be, Mg, and Zn, the incentive being a number of neutron-scattering experiments[10] on the lattice dynamics. Further, a great deal of information is available about the pseudopotential from theoretical studies.[3,4,23] However, to our knowledge, no experimental results have been reported so far for the phonon damping in them. Bearing this in mind our discussions would mostly be based on physical intuitive arguments and on comparison with the phonon frequencies. The calculations for this group are carried out only in the high-symmetry Δ direction [0001] for both the longitudinal- and transverse-acoustic branches. Summation over the reciprocal-lattice vectors is extended to those points for which the energy-wave-number characteristic is significantly different from zero; $6k_F$ in Be and $7k_F$ in Mg and Zn. For this group of metals the SSTL interpolation scheme gives largely deviating values (by an order of magnitude) and hence the results from this scheme are not plotted.

Result are plotted in Fig. 7 only for Be, as a representative of the group because of the similar behavior of the members. However, this time again, the discussions will also be extended to Mg and Zn whenever it seems necessary. A comparison with the phonon frequencies indicates that in Be and Mg the phonon width falls in the range 15–50% of the phonon frequency for different modes and dielectric-screening schemes; whereas in Zn the two are of the same order. The results are reminiscent of the fact that the electron-phonon interaction and the umklapp processes play major roles, while the normal process does not contribute appreciably. This becomes even more striking in Zn in accordance with the experimental findings.[4] There is a steep rise in the damping as we go to higher q values, becoming too high near the zone boundary, which is again a manifestation of the Kohn anomaly and the umklapp process. It is known that employing the free-electron dielectric function cannot be entirely justified for these metals, but this shortcoming has been partly circumvented by using relatively larger values of m^*.

IV. SUMMARY AND CONCLUSIONS

We will finally emphasize a number of general points common to all groups of metals on the basis of the calculations. An important point evident from the results is that the choice of the electron-gas screening function plays a significant role in the electron-phonon interaction. The major difference between the results from different screening schemes generally occurs in longitudinal branches, whereas in transverse branches it oc-

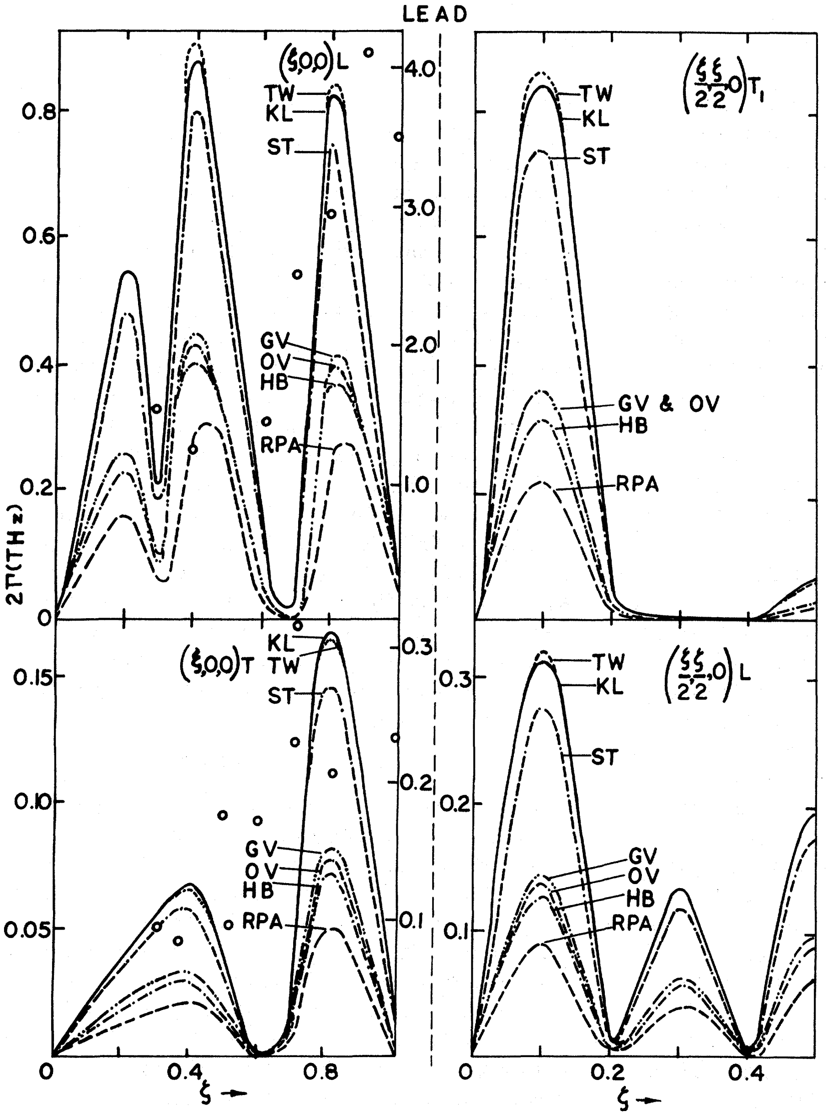

FIG. 6. Same as in Fig. 3 for Pb for (ξ, 0, 0) and (ξ, ξ, 0) directions. The experimental data are sampled from Ref. 9.

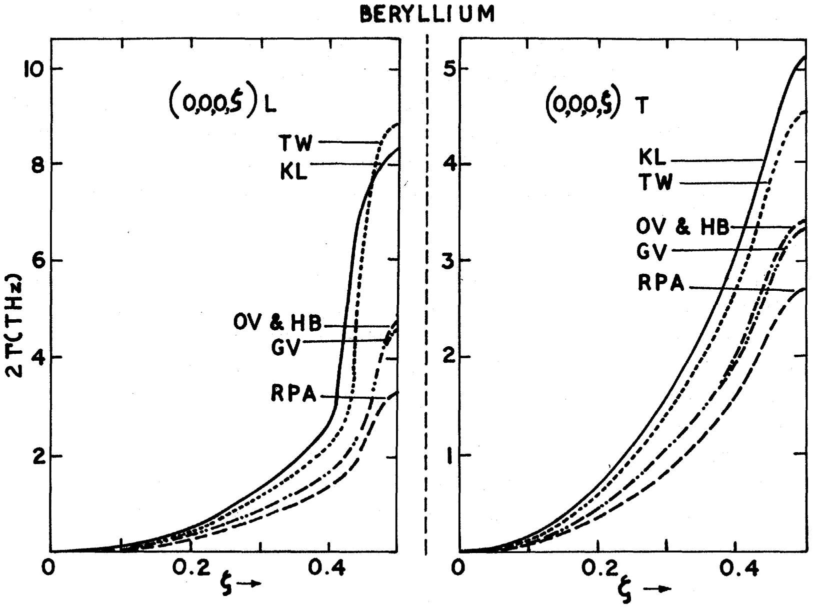

FIG. 7. Same as in Fig. 3 for Be in (0, 0, 0, ζ) direction.

curs only for phonon wave vectors near the zone boundary. This is clear from Fig. 1 as the various $f(q)$'s differ substantially for $q < 2k_F$, i.e., the region of normal process, and tend to be equal for higher values of q, i.e., the region of umklapp process. One, therefore, recalls that until $|\vec{q}|$ gets sufficiently large so as to find a reciprocal-lattice vector h for which $|\vec{q}+\vec{h}| < 2k_F$, the effect of the electron-gas screening function on the transverse branch is not so strong. Further, from a general survey of the results and physical arguments one can categorize the various screening schemes into two groups with RPA, HB, and GV theories falling in one category and SSTL, KL, and TW theories in the other. On the other hand, the values from OV run very close to GV, and as GV has been found to give very good results of the lattice dynamics[25] one expects OV to be a little better than the others. The same thing has also been observed in the electrical resistivity and the thermoelectric power of liquid alkali metals.[32] It therefore seems to be important to calculate the pair correlation function of the electron gas from OV's theory and examine its validity in more detail. The value of the phonon width from the RPA is always the lowest, as it should be, because it completely excludes both the exchange and correlation effects. Conversely, values from KL are the highest most of the time. Unfortunately, from the graphs no prediction regarding the superiority of one dielectric function over the others may be made directly.

A look at the results immediately suggests that the local form of the pseudopotential is a good approximation for most of the elements, but anomalous behavior arises in a number of metals, e.g., Li, Rb, Cs, and Zn, where nonlocality may pose serious difficulties.

It needs to be emphasized that so far we have been concerned only with the phonon damping due to electron-phonon interaction, whereas a more stringent test of the formalism would be via the estimates of the anharmonic contributions where many-body forces would explicitly appear in the pseudopotential for the anharmonic coefficients and then comparing the combined results with the experiments. Another stimulating problem which can further elucidate the pseudoatom formalism and the applicability of the free-electron-assisted dielectric-screening functions seems to be the calculation of the effects of electron-phonon coupling on the infrared properties of metals. This, however, demands the inclusion of the frequency dependence in the screening theory, a proper development which is really an intrincate problem but may prove exceedingly fruitful for studying all the metal properties.

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de Haas–van Alphen Effect in Iridium[*]

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The de Haas–van Alphen effect has been used to study the extremal areas, cyclotron masses and spin-splitting nulls on the two Γ-centered sheets of the Fermi surface of iridium for the magnetic field in a (110) plane. The measured extremal areas are in good agreement with recent relativistic-augmented-plane-wave calculations by Andersen. The mass-enhancement factor determined from a comparison of the calculated and measured masses was found to be 1.31 and rather isotropic for the $\Gamma 6$ sheet; the enhancement on the $\Gamma 5$ sheet is distinctly more anisotropic.

I. INTRODUCTION

The platinum-group transition metals have received a great deal of interest over recent years. Their electronic and magnetic properties have been studied extensively. One characteristic of this group of metals is their high density of states at the Fermi energy, which increases together with the magnetic susceptibility in the sequence Ir, Rh, Pt, and Pd. However, they are unusual in that with the increasing tendency towards ferromagnetism, superconductivity is suppressed. Ir, with a transition temperature of 0.1 °K, is the only one of these metals that has proven to become superconducting.

A detailed knowledge of the electronic band structure in the vicinity of the Fermi level is a necessary foundation for treating electronic, magnetic, and superconducting properties in these metals.

The de Haas-van Alphen (dHvA) effect has been used for experimental investigations of the Fermi surface (FS) and effective cyclotron masses in many metals.[1] Pd[2] and Pt[3] have been exhaustively studied with the dHvA effect; also, the FS of Rh is rather well established experimentally.[4,5] From band-structure calculations[6] the FS of Ir is expected to consist of four different sheets not re-