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Calculated point-contact electron-phonon spectral functions for the alkali metals

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The electron-phonon spectral functions α^2F, $\alpha^2F_{\rm tr}$, and α^2F_p and the corresponding coupling parameters λ, $\lambda_{\rm tr}$, and λ_p central to the theories of superconductivity, transport, and point-contact spectroscopy are calculated for the alkali metals. It is found that the frequently made assumption $\lambda_p \sim \lambda$ is a very poor one. For example: $\lambda_p < \lambda/2$ for K, while $\lambda_p > 2\lambda$ for Cs. The dependence of the point-contact spectral function α^2F_p on the orientation of the crystalline axes relative to the contact is investigated. It is found that the very anisotropic lattice dynamics of the alkali metals lead to an anisotropy of only a few percent in λ_p. The anisotropy in α^2F_p at a given frequency can be a few times larger than this, but we find no significant anisotropy in either peak positions or heights.

INTRODUCTION

Theoretical expressions for various properties of metals in which electron-phonon scattering is important are often formulated in terms of electron-phonon spectral functions of the form[1-3]

$$G(\omega)=\frac{1}{(2\pi)^3}\int_{S_F}\frac{dS_{\vec{k}'}}{\hbar v_{\vec{k}'}}\int_{S_F}\frac{dS_{\vec{k}}}{\hbar v_{\vec{k}}}w(\vec{k},\vec{k}')\sum_{\lambda}|g_{\vec{k}',\vec{k},\lambda}|^2\delta(\hbar\omega-\hbar\omega_{\vec{k}'-\vec{k},\lambda})\Big/\int_{S_F}\frac{dS_{\vec{k}}}{\hbar v_{\vec{k}}}\ . \tag{1}$$

In (1), $g_{\vec{k}',\vec{k},\lambda}$ is the matrix element for the scattering of an electron from state $\vec{k}$ to state $\vec{k}'$ with the emission or absorption of a phonon of wave vector $\vec{k}'-\vec{k}$, polarization index λ, and frequency $\omega_{\vec{k}'-\vec{k},\lambda}$; $dS_{\vec{k}}$ and $dS_{\vec{k}'}$ are elements of area on the Fermi surface S_F, and $w(\vec{k},\vec{k}')$ is a geometrical weight factor appropriate to the property under consideration. Three weight factors of interest are

$$w_s(\vec{k},\vec{k}')=1\ , \tag{2a}$$

$$w_{\rm tr}(\vec{k},\vec{k}')=1-\cos\theta_{\vec{k},\vec{k}'}\ , \tag{2b}$$

$$w_p(\vec{k},\vec{k}')=\frac{1}{2}\left[1-\frac{\theta_{\vec{k},\vec{k}'}}{\tan\theta_{\vec{k},\vec{k}'}}\right]\ , \tag{2c}$$

where $\theta_{\vec{k},\vec{k}'}$ is the angle between $\vec{k}$ and $\vec{k}'$. 2(a) is important in superconductivity,[1] 2(b) in optical properties,[2] 2(a) and 2(b) in transport,[2] and 2(c) in point-contact spectroscopy.[3] These weights factors have been normalized so that they all lead to the same value for

$$\int_0^{\pi}\int_0^{2\pi}\int_0^{\pi}\int_0^{2\pi}w(\vec{k},\vec{k}')\sin\theta_{\vec{k}}d\theta_{\vec{k}}d\phi_{\vec{k}}\times\sin\theta_{\vec{k}'}d\theta_{\vec{k}'}d\phi_{\vec{k}'}\ .$$

Point-contact spectroscopy, which requires the use of the last weighting factor, has the potential to become an important new probe of the electron-phonon interaction in metals (for a recent review, see Ref. 3). However, we believe that results of work to date have been confused by some unjustified assumptions concerning the relationship between the $G(\omega)$'s for these three weighting functions. In this paper we attempt to illustrate this point by presenting detailed calculations for the alkali metals, whose electron-ion interactions are known with considerable accuracy.

The last two weight factors in Eq. (2) do not take into account Fermi-surface distortions or anisotropy in the electron-phonon interaction. If the latter restriction is removed, the point-contact weight factor becomes[3]

$$w_p(\vec{k},\vec{k}')=4\left[\left(\frac{k_x}{k_z}-\frac{k_x'}{k_z'}\right)^2+\left(\frac{k_y}{k_z}-\frac{k_y'}{k_z'}\right)^2\right]^{-1/2}\times\Theta(-k_zk_z')\ , \tag{3}$$

where the coordinate system has been chosen so that the origin coincides with the center of the (circular) region of contact and $\hat{z}$ is normal to the plane of contact. The orientation of the crystallographic axes, specified by the unit vectors $\hat{i}$, $\hat{j}$, and

$\hat{k}$, with respect to the contact, is expressed by the direction cosines $\gamma_i \equiv \hat{i}\cdot\hat{z}$, $\gamma_j \equiv \hat{j}\cdot\hat{z}$, and $\gamma_k \equiv \hat{k}\cdot\hat{z}$.

The electron-phonon spectral functions $G(\omega)$ corresponding to the weight factors (2a), (2b), and (2c) are customarily denoted by $\alpha^2F(\omega)$, $\alpha^2F_{\mathrm{tr}}(\omega)$, and $\alpha^2F_p(\omega)$; that corresponding to (3) will be denoted by $\alpha^2F_p(\omega;\gamma_i,\gamma_j,\gamma_k)$ in this paper. For a given spectral function $G(\omega)$ the quantity

$$2\int_0^\infty d\omega \frac{G(\omega)}{\omega} \tag{4}$$

often provides a useful measure of the effective strength of the electron-phonon interaction. The electron-phonon coupling parameters obtained by substituting $\alpha^2F(\omega)$, $\alpha^2F_{\mathrm{tr}}(\omega)$, $\alpha^2F_p(\omega)$, and $\alpha^2F_p(\omega;\gamma_i,\gamma_j,\gamma_k)$ for $G(\omega)$ in (4) are denoted by λ, λ_{tr}, λ_p, and $\lambda_p(\gamma_i,\gamma_j,\gamma_k)$.

It is frequently assumed that $\alpha^2F_p(\omega)$ is not very different from $\alpha^2F(\omega)$ and consequently that $\lambda_p \approx \lambda$.[3,4] In fact, van Gelder[5] has proven that $\lambda = \lambda_{\mathrm{tr}} = \lambda_p$ for a simple model in which $|g|^2 \propto \omega$. Another assumption is that the anisotropy in the point-contact spectral function of a cubic metal is negligible so that $\alpha^2F_p(\omega;\gamma_i,\gamma_j,\gamma_k)$ is only very weakly dependent on the orientation of the crystal axes and, hence, equal to $\alpha^2F_p(\omega)$ to a very good approximation.[3] In this paper we show that the first of these assumptions is not valid in the alkali metals.

II. CHOICE OF INPUT TO CALCULATIONS

In what follows we assume that the Fermi surface is spherical and accordingly use the one-OPW (orthogonalized-plane-wave) result[6]

$$|g_{\vec{k}',\vec{k},\lambda}|^2 = \frac{\hbar[(\vec{k}'-\vec{k})\cdot\vec{e}_{\vec{k}'-\vec{k},\lambda}]^2 V^2(|\vec{k}'-\vec{k}|)}{2MN\omega_{\vec{k}'-\vec{k},\lambda}} \tag{5}$$

for the electron-phonon coupling. Although this is only well justified for Na, K, and Rb, the qualitative conclusions of this paper should apply for Li and Cs as well. In (5), $\vec{e}_{\vec{k}'-\vec{k},\lambda}$ is a phonon polarization vector, V is the screened electron-ion pseudopotential form factor for scattering at the Fermi surface, M is the ionic mass, and N is the number ions per unit volume.

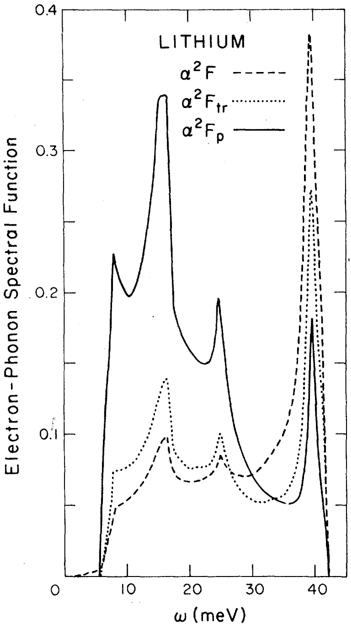

FIG. 1. Calculated electron-phonon spectral functions $\alpha^2F(\omega)$, $\alpha^2F_{\mathrm{tr}}(\omega)$, and $\alpha^2F_p(\omega)$ for Li (lattice constant is 3.478 Å).

Since the detailed results are quite sensitive to the choice of input to the calculations, we have used the most recent refinement[7] of the first-principles lattice dynamics $(\omega,\vec{e})$ and electron-ion interaction (V) developed originally by Dagens, Rasolt, and Taylor.[8] The bare electron-ion pseudopotential was determined by fitting the linear-response calculation of the charge density induced about an isolated ion embedded in an electron gas to Dagens's full nonlinear calculation of the same

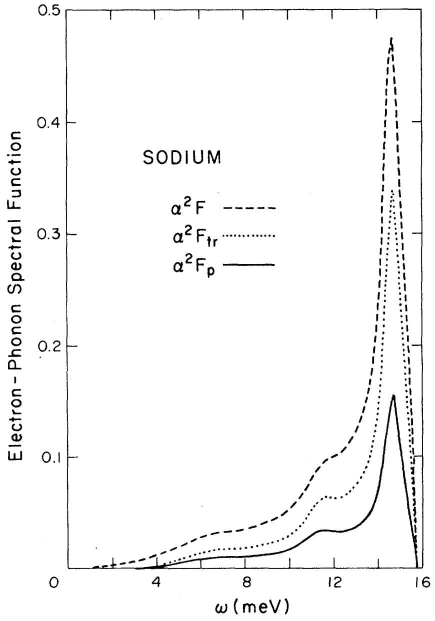

FIG. 2. Calculated electron-phonon spectral functions $\alpha^2F(\omega)$, $\alpha^2F_{\mathrm{tr}}(\omega)$, and $\alpha^2F_p(\omega)$ for Na (lattice constant is 4.225 Å).

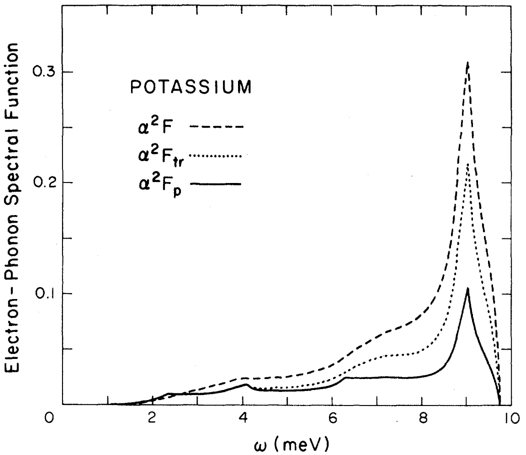

FIG. 3. Calculated electron-phonon spectral functions $\alpha^2F(\omega)$, $\alpha^2F_{\text{tr}}(\omega)$, and $\alpha^2F_p(\omega)$ for K (lattice constant is 5.2355 Å).

quantity. This pseudopotential used in combination with the dielectric function of Geldart and Taylor[9] and the Rasolt form[10] for the vertex correction[11] has proven very successful in calculations of various properties[12] for Na, K, and Rb, giving us considerable confidence in the present results for these metals. We do not believe that variation of our choice of input within reasonable bounds (i.e., which does not destroy the good *quantitative* agreement with experiment already obtained for several other properties[12]) can possibly change the *qualitative* conclusions of this paper.

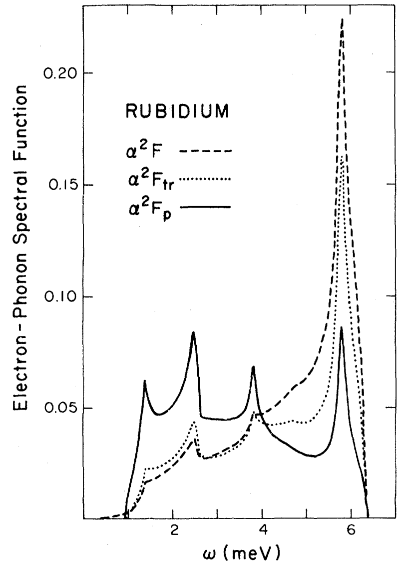

FIG. 4. Calculated electron-phonon spectral functions $\alpha^2F(\omega)$, $\alpha^2F_{\text{tr}}(\omega)$, and $\alpha^2F_p(\omega)$ for Rb (lattice constant is 5.5850 Å).

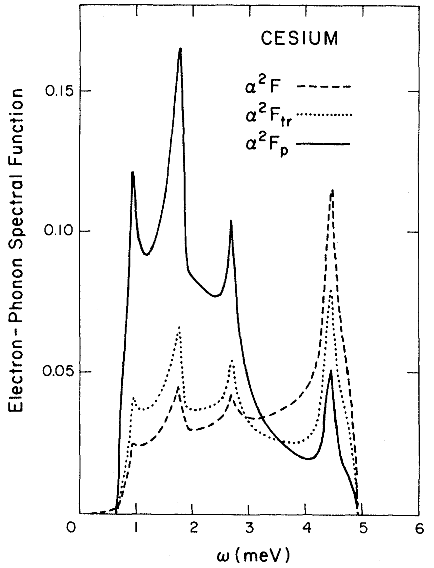

FIG. 5. Calculated electron-phonon spectral functions $\alpha^2F(\omega)$, $\alpha^2F_{\text{tr}}(\omega)$, and $\alpha^2F_p(\omega)$ for Cs (lattice constant is 6.045 Å).

III. NUMERICAL RESULTS AND DISCUSSION

In Figs. 1–5 our calculated results for $\alpha^2F(\omega)$, $\alpha^2F_{\text{tr}}(\omega)$, and $\alpha^2F_p(\omega)$ are shown for each of Li, Na, K, Rb, and Cs. The corresponding coupling parameters λ, λ_{tr}, and λ_p are given in Table I. Obviously, the assumption that $\alpha^2F_p(\omega)$ is not very different from $\alpha^2F(\omega)$ is a poor one as is the expectation that $\lambda \approx \lambda_p$. The worst cases are for Na where $\lambda_p < \lambda/3$ and for Cs where $\lambda_p > 2\lambda$. Hence comparison[3,4] of experimental values of λ_p with theoretical values of λ is not a meaningful test of either the experimental or theoretical results, nor can point-contact spectroscopy be used, as has been claimed,[3] to measure directly the mass-enhancement parameter λ. Table I also contains a comparison of two sets of experimental results for λ_p with each other and with our calculated values. It is somewhat discouraging that there is no metal, except possibly Na, for which there is good agreement between all three values of λ_p.

Table II contains our calculated values of $\lambda_p(\gamma_i,\gamma_j,\gamma_k)$ for three choices of the direction cosines $(\gamma_i,\gamma_j,\gamma_k)$ corresponding, respectively, to a crystal oriented such that the [100], [110], and [111] directions (on both sides of the contact) are normal to the contact plane. It is seen that the very considerable anisotropy in the electron-phonon interaction has been almost completely averaged out leaving a $\lambda_p(\gamma_i,\gamma_j,\gamma_k)$ with an anisotropy of only a few percent. It is also found that

TABLE I. A comparison, for the bcc alkali metals, of the calculated electron-phonon coupling parameters λ^{calc}, $\lambda_{tr}^{\text{calc}}$, and λ_p^{calc} with each other and with the experimental results for λ_p presented in Refs. 3 and 4.

Metal	λ^{calc}	$\lambda_{\text{tr}}^{\text{calc}}$	λ_p^{calc}	Expt. Ref. 3	Expt. Ref. 4
Li	0.301	0.316	0.665	0.45 ± 0.20	0.25 ± 0.10
Na	0.202	0.117	0.060	0.10 ± 0.03	0.064 ± 0.012
K	0.136	0.086	0.058	0.13 ± 0.03	0.13 ± 0.02
Rb	0.141	0.123	0.165		
Cs	0.128	0.141	0.294		

$\alpha^2F_p(\omega;\gamma_i,\gamma_j,\gamma_k)$ for a given crystal orientation does not differ from $\alpha^2F_p(\omega)$ by a frequency-independent scale factor and that the anisotropy in $\alpha^2F_p(\omega;\gamma_i,\gamma_j,\gamma_k)$ at a given frequency can be up to a few times larger than that in the integrated quantity $\lambda_p(\gamma_i,\gamma_j,\gamma_k)$. However, as shown for Rb in Fig. 6, we do not find any significant dependence on orientation of either peak positions or heights. Hence, bearing in mind the thermal smearing of the experimentally observed spectral functions, the assumption[3] that the anisotropy in α^2F_p is negligible for cubic metals is completely borne out by our calculations. As predicted,[3] the very considerable anisotropy of the lattice dynamics is almost completely washed out by the double Fermi-surface integral over $\vec{k}_F$ and $\vec{k}_F'$. Our calculations do not, however, allow for anisotropy arising from either Fermi-surface distortions or noncircular point contacts. In particular, they do not rule out considerable anisotropy in the case of cubic transition metals.

It should be noted that there is a qualitative relationship between the results reported in Tables I and II and the size of the electron-ion form factor for scattering across the Fermi surface, i.e., $|V(2k_F)/V(0)|$. (The values of this quantity for Li, Na, K, Rb, and Cs are 0.273, 0.025, 0.057, 0.172, and 0.272, respectively.[12]) Table I shows that $\lambda_p<\lambda$ for Na and K with small values of $|V(2k_F)/V(0)|$ and $\lambda_p>\lambda$ for Li, Rb, and Cs with much larger values. This is not surprising when one considers the ratio $w_p(\vec{k},\vec{k}')/w_s(\vec{k},\vec{k}')$ of the relevant weight factors in the $|\vec{k}'-\vec{k}|\to0$ and $|\vec{k}'-\vec{k}|\to2k_F$ limits (0 and ∞, respectively). Clearly, if the integrand exclusive of the weight factor $w(\vec{k},\vec{k}')$ is large (small) enough near $|\vec{k}'-\vec{k}|=2k_F$ compared to what it is for $|\vec{k}'-\vec{k}|\ll2k_F$ then λ_p will be greater (less) than λ. Apart from simple scaling the material dependence of this part of the integrand is almost entirely contained in $V(|\vec{k}'-\vec{k}|)$; hence the importance of $V(2k_F)/V(0)$ in comparing qualitative differences such as the above among the alkali metals.

Because of the singular behavior of $w_p(\vec{k},\vec{k}')$ for $\vec{k}'-\vec{k}=2\vec{k}_F$, it is interesting and instructive to consider the following two density-of-states functions for $2\vec{k}_F$ phonons: $f(\omega)d\omega$ is the fraction of $2\vec{k}_F$ phonons encountered in computing (1) with frequency between ω and $\omega+d\omega$; $f_{\text{eff}}(\omega)d\omega$, the effective fraction of $2\vec{k}_F$ phonons, is obtained by associating with each such phonon a weight factor

$$(2\vec{k}_F\cdot\vec{e}_{2\vec{k}_F,\lambda})^2/\omega_{2\vec{k}_F,\lambda}\ .$$

Figure 7 shows $f(\omega)$ and $f_{\text{eff}}(\omega)$ for Rb. It is seen that the factors $\omega_{2\vec{k}_F,\lambda}^{-1}$, and especially $(2\vec{k}_F\cdot\vec{e}_{2\vec{k}_F,\lambda})^2$, dramatically reduce $f_{\text{eff}}(\omega)$ at high frequencies so that the effective number of $2\vec{k}_F$ phonons in the region of the longitudinal peak is very small, much less than for the lower frequency peaks. This allows the longitudinal peak in $\alpha^2F(\omega)$

TABLE II. Anisotropy of the point-contact electron-phonon coupling parameter $\lambda_p(\gamma_i,\gamma_j,\gamma_k)$ for the bcc alkali metals.

Metal	$\lambda_p(1,0,0)/\lambda_p$	$\lambda_p(2^{-1/2},2^{-1/2},0)/\lambda_p$	$\lambda_p(3^{-1/2},3^{-1/2},3^{-1/2})/\lambda_p$
Li	1.022	0.997	0.984
Na	1.010	1.000	0.993
K	1.013	0.999	0.992
Rb	1.019	0.999	0.988
Cs	1.021	0.997	0.984

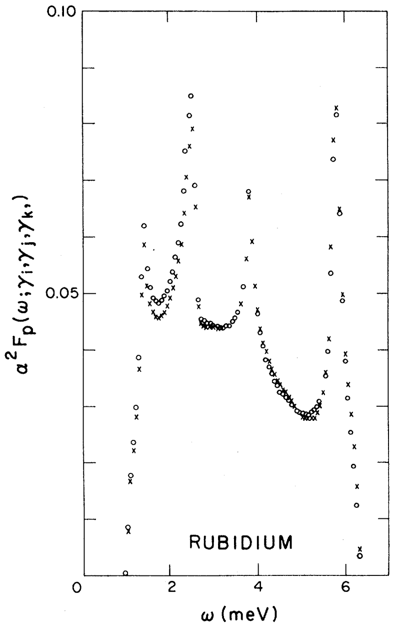

FIG. 6. Anisotropic point-contact spectral function $\alpha^2F_p(\omega;\gamma_i,\gamma_j,\gamma_k)$ of Rb for two different choices of the direction cosines $\gamma_i,\gamma_j,\gamma_k$. The symbols ○ and × denote $(\gamma_i,\gamma_j,\gamma_k)=(1,0,0)$ and $(3^{-1/2},3^{-1/2},3^{-1/2})$, respectively.

to remain larger than that in $\alpha^2F_p(\omega)$ for all the alkali metals despite the (integrable) singularity at $2\vec{k}_F$ contributing to the latter spectral function. The dominant contribution to the longitudinal peak of $\alpha^2F(\omega)$ evidently comes from phonons far from the singularity where $w_s(\vec{k},\vec{k}')>w_p(\vec{k},\vec{k}')$.

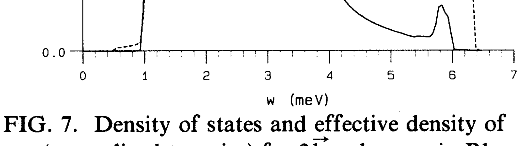

FIG. 7. Density of states and effective density of states (normalized to unity) for $2\vec{k}_F$ phonons in Rb. $f(\omega)d\omega$ is the fraction of $2\vec{k}_F$ phonons with frequency between ω and $\omega+d\omega$ encountered in computing the various electron-phonon spectral functions $G(\omega)$. $f_{\text{eff}}(\omega)d\omega$ is the effective fraction of $2\vec{k}_F$ phonons obtained by associating with each such phonon the weight factor $(2\vec{k}_F\cdot\vec{e}_{2\vec{k}_F,\lambda})^2/\omega_{2\vec{k}_F,\lambda}$.

When $|V(2k_F)/V(0)|$ is very large, as in Li and Cs, the low-frequency peaks in $\alpha^2F_p(\omega)$ with their relatively much larger values of $f_{\text{eff}}(\omega)$ dominate the higher-frequency longitudinal peak. The latter is too high in frequency to contain a significant contribution from $2\vec{k}_F$ phonons.

It is important to note that conclusions about the relative strengths of the couplings to high-frequency and low-frequency phonons drawn from point-contact measurements could be in error if one assumed $\alpha^2F\equiv\alpha^2F_p$. For example, the interpretation[3] of Li data along these lines is somewhat misleading (see Fig. 1), and similar conclusions for for the noble metals[3] should be regarded with some caution.

In the alkali metals, the most anisotropic feature of the phonon dispersion is the anomalously low T_1 branch in the [110] direction and the minimum wave-vector transfer for an umklapp process occurs for $|\vec{k}'-\vec{k}|=2k_F$ with $\vec{k}'-\vec{k}$ in the [110] direction. Moreover, the integral defining λ_p gives preferential weight to the low-frequency part of α^2F_p both because of the explicit factor of ω^{-1} in Eq. (4) and the frequency dependence of $f_{\text{eff}}(\omega)$. Hence, one expects the anisotropy in λ_p to be larger the larger the value of $|V(2k_F)/V(0)|$, as observed in Table II. Similarly, the onset of the umklapp processes seen in Figs. 1–5 is more pronounced when $|V(2k_F)/V(0)|$ is larger. When this quantity is very large, as in Li and Cs, the onset is quite dramatic.

We should emphasize that our calculations have not included the influence of distortions of the electron dispersion from the free-electron parabola. This approximation should not be serious for Na, K, or Rb but it could be significant in Li and Cs which are known to have band masses larger than the free-electron value.[13] Since each of the λ values contains a net factor of the Fermi-level density of states, these large band masses might be expected to increase the λ values. However, a larger Fermi-level density of states will also increase the screening of the electron-ion interaction and thereby reduce $|g_{\vec{k}',\vec{k},\lambda}|^2$ [see Eq. (5)]. Using the comparison of calculated and measured electrical resistivities[14,15] at temperatures near the respective Debye temperatures as a guide, we estimate that band effects should increase λ by ~30% in Li and by ~20% in Cs.

IV. CONCLUDING REMARKS

In summary, we find that there is no simple relationship between electron-phonon spectral functions calculated with different weight factors in the alkali metals. Since there is no reason to expect the situation to be different in other systems, we believe that the value of point-contact spectroscopy in probing electron-phonon interactions in transition metals and other systems will be greatly enhanced if theoretical calculations of the same quantity are undertaken. We believe such a comparison will be useful in systems in which the electron-phonon interaction is not as accurately known, even if tunnelling data already exists.

Since submitting this paper we learned of prior calculations of the point-contact spectral functions of Na and K by Ashraf and Swihart.[16] It is difficult to make a detailed comparison of their calculated spectral functions with ours because of the numerical noise in their curves. Moreover, none of the figures in their paper are directly comparable to any of ours because they are either for different electron-ion form factors or different temperatures. (All our calculations are for $T=0$.) However, the qualitative agreement is entirely satisfactory. Nevertheless, our conclusions are very different from those of Ashraf and Swihart. They have emphasized that α^2F and α^2F_p are similar in shape even though there is a substantial disagreement in overall magnitude. It is apparent from our more extensive calculations that this is not a general property of the alkali metals: The shape similarity found for Na and K does not extend to the other alkali metals. We emphasize again that there is no simple relationship between α^2F and α^2F_p and caution against any conclusions drawn on the basis of an expected similarity.

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