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On calculating the electron-phonon mass enhancement λ for compounds

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Abstract. We present a technique for calculating the electron mass-enhancement factor λ for compounds which should be particularly accurate for binary materials where one constituent is much heavier than the other. We find that λ decomposes into a sum of terms from each independent site in the unit cell. Each term involves an electron-phonon interaction factor which may be evaluated by the method of Gaspari and Gyorffy; and a phonon spectrum moment which is weighted by the amplitude of vibration of the atom in question. The contribution to λ from the heavy atom involves electron coupling to the acoustic phonons, while the light atom couples to the optic phonons. A comparison of our calculations for the refractory carbides and the palladium hydrogen system indicates that our proposed formulation gives reliable results.

1. Introduction

Recent theoretical work (Gaspari and Gyorffy 1972, Evans *et al* 1973, Gomersall and Gyorffy 1974) has made it possible to do relatively accurate calculations of the parameters that govern the superconducting properties of elements and some compounds. (See for instance, Evans *et al* 1973, Klein and Papaconstantopoulos 1974 and 1975, Papaconstantopoulos and Klein 1975, Gomersall and Gyorffy 1974.) These calculations make use of two ingredients: (i) *ab initio* electronic band structure calculations (by say the APW or KKR methods) to compute the electron-phonon interaction (EPI), and (ii) the measured phonon density of states (usually by neutron scattering). Following the ideas of McMillan (1968), the electron mass enhancement factor λ can then be calculated with the assumption that the tunneling density of states, $\alpha^2(\omega) F(\omega)$, is proportional to the phonon density of states, $F(\omega)$, over the whole phonon frequency range. For monatomic materials this appears to be a good approximation.

For the case of polyatomic compounds, where there are both acoustic and optic phonon modes, the validity of the latter approximation is questionable, and there remains the question of how to accurately evaluate λ . This is the main point which we address in this paper. In particular, we will derive and discuss a specific extension of the McMillan equations for λ for compounds which should be especially accurate for binary materials where one of the constituents is much heavier than the other. A number

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of interesting materials fall into this category, such as, the refractory carbides (NbC, TaC, HfC, etc.), and the palladium–hydrogen system (PdH and PdD).

The plan of the paper is as follows: In §2, we first review the McMillan theory for monatomic materials and then derive an extension of the theory for compounds; and in §3 we compare our approach with that of other workers. In §4 we compare the calculations that have been done for compounds with the available experimental information, and we show that our theoretical approach is in good agreement with these experiments. We then present a summary in §5.

2. Theory

In the strong coupling theory of superconductivity (Eliashberg 1960, 1961 and McMillan 1968), the fundamental quantity to determine is the function $\alpha^2(\omega) F(\omega)$, where $F(\omega)$ is the phonon density of states, and $\alpha^2(\omega)$ is related to an average of the EPI. Explicitly we have (all quantities in atomic units)

$$\alpha^2(\omega) F(\omega) = \frac{1}{(2\pi)^3} \sum_{\nu} \left(\int \int d^3k d^3k' |g_{\mathbf{k},\mathbf{k}',\nu}|^2 \delta(E_{\mathbf{k}} - E_{\mathbf{k}'}) \right. \\ \left. \times \delta(E_{\mathbf{k}} - E_{\mathbf{k}'}) \delta(\omega - \omega_{\mathbf{k}-\mathbf{k}',\nu}) \right) / \int d^3k \delta(E_{\mathbf{k}} - E_{\mathbf{F}}) \quad (1)$$

with $E_{\mathbf{k}}$ denoting an electronic energy, $\omega_{\nu}(\mathbf{q})$ a phonon energy in mode ν , and $E_{\mathbf{F}}$ the Fermi energy. The EPI is contained in $g_{\mathbf{k}\mathbf{k}',\nu}$ which is given in the rigid ion approximation by

$$g_{\mathbf{k}\mathbf{k}',\nu} = \sum_{s=1}^{N_s} \left(\frac{1}{2M_s \Omega \omega_{\nu}(\mathbf{k} - \mathbf{k}')} \right)^{1/2} \left(\int d^3r \psi_{\mathbf{k}}^*(\mathbf{r}) (\mathbf{e}(\mathbf{k} - \mathbf{k}'|\nu s) \cdot \nabla V_s) \psi_{\mathbf{k}}(\mathbf{r}) \right). \quad (2)$$

The label s is for the N_s atoms in the unit cell, having masses M_s , and phonon polarization vectors $\mathbf{e}(\mathbf{q}|\nu s)$. $V_s(\mathbf{r})$ is the standard muffin-tin potential which gives the eigenenergies and eigenfunctions $E_{\mathbf{k}}$ and $\psi_{\mathbf{k}}$, and the integral is over a unit cell of volume Ω .

The fundamental parameter λ is defined by

$$\lambda = 2 \int \alpha^2(\omega) F(\omega) d\omega / \omega \quad (3)$$

and it was first noted by McMillan (1968), that the first moment of $\alpha^2 F$ is independent of phonon frequencies, and is given by

$$\int \omega \alpha^2(\omega) F(\omega) d\omega = n(E_{\mathbf{F}}) \sum_{n=1}^{N_s} \langle I_s^2 \rangle / 2M_s. \quad (4)$$

Gaspari and Gyorffy (1972) showed how $\langle I_s^2 \rangle$ is related to electronic parameters by the expression (in atomic units),

$$\langle I_s^2 \rangle = \frac{E_{\mathbf{F}}}{\pi^2 n^2(E_{\mathbf{F}})} \sum_l \frac{2(l+1) \sin^2(\delta_{l+1,s} - \delta_{l,s}) n_{l,s} n_{l+1,s}}{n_{l,s}^{(1)} n_{l+1,s}^{(1)}}. \quad (5)$$

The $\delta_{l,s}$ are scattering phase shifts, $n_{l,s}$ are the site and angular momentum decomposed densities of states and the $n_{l,s}^{(1)}$ are 'free-scatterer' densities of states; all

quantities being discussed fully in the paper by Evans *et al* (1973) and Gomersall and Gyorffy (1974). The derivation leading to equation (5) neglects small 'off-diagonal' scattering terms (B L Gyorffy, private communication). Equation (4) is obtained by making use of the closure relation

$$\sum_v e_i^*(\mathbf{q}|vs) e_j(\mathbf{q}|vs') = \delta_{s,s'} \delta_{i,j} \quad (6)$$

in equations (1) and (2), with (i, j) labelling the cartesian components of e .

For monatomic materials ($N_s = 1$), it has been found from tunneling measurements that, in general, $\alpha^2(\omega)F(\omega)$ has very much the same shape as $F(\omega)$ itself, so that the moments of $\alpha^2(\omega)F(\omega)$ are to a good approximation the same as the moments of $F(\omega)$ (both distributions being properly normalized). That is, for the purpose of evaluating moments, $\alpha^2(\omega)$ can be treated as a constant. With this assumption, and using equations (3) and (4), McMillan (1968) showed that

$$\lambda = \frac{n\langle I^2 \rangle}{M \overline{\omega^2}} \quad (7)$$

with

$$\overline{\omega^2} = \langle \omega \rangle / \langle \omega^{-1} \rangle \quad (8)$$

$$\langle \omega^n \rangle = \int_0^\gamma \omega^n F(\omega) d\omega \quad (9)$$

and

$$\langle \alpha^2 \rangle = \frac{n\langle I^2 \rangle}{2M \langle \omega \rangle}. \quad (10)$$

This approximation for monatomic materials is not likely to be very serious, and is, of course very convenient, as λ separates into an 'electronic part' calculated from first principles, and a 'phonon part' at present obtained from experiment (neutron scattering): numerator and denominator of equation (7).

For the case of compounds, the assumption of constant $\alpha^2(\omega)$ over the whole phonon frequency range is likely to be very poor. This is due to the fact that different frequency intervals correspond to phonon modes in which the displacement of one atom or another is emphasized; and since the electronic states associated with the various atoms may be very different (for instance a compound between a transition metal and a non-transition metal), one would expect that the EPI coupling may be strongly frequency dependent. However, since the direct evaluation of equation (1) is currently a formidable task, one would like to obtain an accurate generalization of equation (7) for compounds. For the case of a binary compound where one atom is much heavier than the other, we now show how this can be achieved with good accuracy.

Suppose in the binary compound, $M_1 \gg M_2$, then to a good approximation (see Appendix), we may write for the bulk of the Brillouin zone:

$$\begin{aligned} e(\mathbf{q}|v2) &\cong 0 && v \text{ acoustic} \\ e(\mathbf{q}|v1) &\cong 0 && v \text{ optic.} \end{aligned} \quad (11)$$

Furthermore, there will be an energy gap between the acoustic and optic modes, so that we may speak of separate acoustic and optic mode frequency regions. Under these conditions, we introduce the key approximation

$$\alpha^2(\omega) = \begin{cases} \alpha_1^2 & \omega \leq \omega_1 \\ \alpha_2^2 & \omega \geq \omega_2 \end{cases} \tag{12}$$

where α_1^2 and α_2^2 are assumed constant, and ω_1 represents the cutoff of the acoustic modes, while ω_2 is the lowest optic mode frequency.

Making use of equations (1), (2), (11) and (12) we arrive at

$$\alpha_1^2 \int_0^{\omega_1} \omega F(\omega) d\omega = n \langle I_1^2 \rangle / 2M_1$$

$$\alpha_2^2 \int_{\omega_2}^{\infty} \omega F(\omega) d\omega = n \langle I_2^2 \rangle / 2M_2$$

or

$$\alpha_s^2 = \frac{n \langle I_s^2 \rangle}{2M_s \langle \omega \rangle_s} \quad s = 1, 2 \tag{13a}$$

and

$$\lambda = \lambda_1 + \lambda_2 \tag{13b}$$

$$\lambda_s = \frac{n \langle I_s^2 \rangle}{M_s \omega_s^2} = 2\alpha_s^2 \langle \omega^{-1} \rangle_s$$

with

$$\langle \omega^n \rangle_s = \begin{cases} \int_0^{\omega_1} \omega^n F(\omega) d\omega & s = 1 \\ \int_{\omega_2}^{\infty} \omega^n F(\omega) d\omega & s = 2 \end{cases} \tag{13c}$$

and

$$\overline{\omega_s^2} = \langle \omega \rangle_s / \langle \omega^{-1} \rangle_s. \tag{13d}$$

Note that we have chosen the normalization condition

$$\int_0^{\omega_1} F(\omega) d\omega = \int_{\omega_2}^{\infty} F(\omega) d\omega = 1.$$

3. Comparison with other approaches

Equations (13) are our central results, and equation (13b) is similar to a relation proposed by Phillips (1972) in his discussion of the superconducting properties of the refractory carbides and nitrides. There is an important difference however, in that Phillips' equivalent to our equation (13b) has numerators: $n_s \langle I_s^2 \rangle$ where n_s is the density of states at E_F associated with site s . The formalism that we have outlined

gives our equations (5) and (13b) and indicates the approximate nature of Phillips' expression.

The important step leading to our results in equation (13) is the assumption embodied in our equation (12) which allows the electrons freedom to couple differently to the acoustic and optic modes. This is likely to be a good approach for treating general compounds, but the final results, equations (13a, b), explicitly make use of the condition that the $M_1 \gg M_2$, and follows from neglecting terms of order (M_2/M_1) .

In two papers discussing the EPI in NbN and NbC, Gomersall and Gyorffy (1973, 1974) proposed the formula

$$\lambda_s = \frac{n \langle I_s^2 \rangle}{M_s \overline{\omega^2}} \quad (14)$$

where $\overline{\omega^2}$ is an average over the whole phonon spectrum acoustic and optic modes. From our previous discussion, it can be seen that one would arrive at this result by assuming a single constant α^2 over the whole frequency range. In their first paper Gomersall and Gyorffy (1973), choose $\overline{\omega^2}$ by using an unweighted average over the phonon density of states, while in their second paper (Gomersall and Gyorffy 1974), they attempt to improve things by weighting $\alpha_s^2 = \text{constant}/\langle \omega \rangle_s$. Although this form for α_s^2 is the same as our equation (13a), Gomersall and Gyorffy still used equation (14) for λ , with the same $\overline{\omega^2}$ for both sites. We have shown that once the restriction that $\alpha_1^2 = \alpha_2^2$ is removed, and $M_1 \gg M_2$, the correct form for λ is given in equation (13). We will show below that the use of equation (14) can lead to substantial qualitative errors.

4. Calculations and comparison with experiment

In table 1 we summarize calculations that we have done for NbC and TaC (Klein and Papaconstantopoulos 1974) and PdD (Papaconstantopoulos and Klein 1975) for which the condition $M_1 \gg M_2$ holds reasonably well. All of these materials have the sodium chloride structure and have superconducting transition temperatures, $T_c \approx 10$ K.

Table 1. Calculated values of λ and α^2 for NbC, TaC and PdD using equations (13a-d).

		λ	α^2 (K)
NbC ^a	{Nb	0.45}	0.63
	{C	0.18}	
TaC ^a	{Ta	0.47}	0.66
	{C	0.19}	
PdD ^b	{Pd	0.21}	0.93
	{D	0.72}	

^a (Klein and Papaconstantopoulos 1974)

^b (Papaconstantopoulos and Klein 1975).

However, we note from table 1 that the contributions to λ are mainly from the transition metal site for the carbides, but is primarily from the deuterium site in PdD. That is, in the former materials, acoustic mode coupling is dominant, while in the latter, coupling to the optic modes gives the major contribution to λ . The strongest confirmation of our use of equation (13) comes from two recent sets of experiments on NbC and on PdH and PdD which we now discuss separately.

Superconducting tunneling measurements on NbC by Geerk *et al* (1975) give an acoustic mode contribution to λ of 0.51 which is close to our calculated value of 0.45 based on equation (13). Experimental difficulties in doing tunneling measurements in the high frequency optic mode region (~ 70 meV) of NbC have prevented an explicit measurement of λ_{optic} . However, since the empirical value of the total λ for NbC is 0.61 (using the measured T_c and the McMillan (1968) equation), it must be that λ_{optic} is much smaller than $\lambda_{\text{acoustic}}$, as we have obtained in our calculations. If we had used the Gomersall and Gyorfy form for λ_s , equation (14), we would have underestimated $\lambda_{\text{acoustic}}$ by more than a factor of two.

Tunneling measurements on PdD and PdH by Dynes and Garno (1975) indicate that the major contribution to λ is coming from the optic modes, in agreement with our calculations on PdD (our calculated results for λ for PdH will be nearly the same, with possibly a small difference in λ for the two isotopes; see Papaconstantopoulos and Klein (1975) and Klein and Papaconstantopoulos (1975)). Somewhat conflicting tunneling measurements by Eichler *et al* (1975) show approximately equivalent contributions from the acoustic and optic modes, while tunneling measurements by Silverman and Briscoe (1975) seem to agree with Dynes and Garno (1975). In addition, resistivity measurements by McLachlan *et al* (1975) show that $\lambda_{\text{optic}} \approx 3 \lambda_{\text{acoustic}}$ in very good agreement with our calculations.

5. Conclusions

In summary, the experimental evidence lends very good support to our use of equations (13) for binary compounds where $M_1 \gg M_2$. We not only obtain relatively accurate values of the total λ , but we also find that the relative contributions from the acoustic modes (heavy atom) and optic modes (light atom) appear to be correct. We hope that the tunneling measurements can be accurately extended to the high frequency region so that we can, in addition, check our calculated values of (α_1^2/α_2^2) and λ_{optic} .

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Appendix

In this appendix we present a somewhat detailed justification of the assertion equation (11) in the text.

We begin by writing the dynamical matrix in the standard form (Maradudin *et al* 1971)

$$\mathbf{D}_{ij}(ss'|\mathbf{q}) = (M_s M_{s'})^{-1/2} \sum_l \Phi_{ij}(l s; l' s') \exp\{i\mathbf{q} \cdot [\mathbf{x}(l) - \mathbf{x}(l')]\} \quad (\text{A.1})$$

with,

$$\Phi_{ij}(l s; l' s') = \left. \frac{\partial^2 \Phi}{\partial U_i(l s) \partial U_j(l' s')} \right|_0$$

the force constant matrix. Here (i, j) are cartesian indices ($i = 1, 2, 3$); s labels the atoms in the unit cells having locations $\mathbf{x}(l)$; Φ is the potential energy; M_s are the masses; and $U(l, s)$ is the displacement of atom (l, s) from equilibrium. The eigenvalues of the dynamical matrix are the phonon frequencies $\omega_v^2(\mathbf{q})$, and the eigenvectors are the phonon polarization vectors $\mathbf{e}(\mathbf{q}|v s)$, with v the branch index. The polarization vectors satisfy the closure relations

$$\begin{aligned} \sum_{s,i} e_i^*(\mathbf{q}|v s) e_i(\mathbf{q}|v' s) &= \delta_{v v'} \\ \sum_v e_i^*(\mathbf{q}|v s) e_j(\mathbf{q}|v s') &= \delta_{ij} \delta_{ss'} \end{aligned} \quad (\text{A.2})$$

We now specialize to the case of two atoms per unit cell ($s = 1, 2$). For this situation it can be shown (Maradudin *et al* 1971) that in the long-wavelength limit, $\mathbf{q} \rightarrow 0$, the following exact result holds

$$\frac{|e(0|v_{ac}2)|}{|e(0|v_{ac}1)|} = \left(\frac{M_2}{M_1} \right)^{1/2} \quad (\text{A.3})$$

and

$$\frac{|e(0|v_{op}1)|}{|e(0|v_{op}2)|} = \left(\frac{M_2}{M_1} \right)^{1/2}$$

where v_{ac} denotes the three acoustic mode branches ($v = 1, 2, 3$) for which $\omega_{v_{ac}}^2(0) = 0$, and v_{op} are the three optic mode branches ($v = 4, 5, 6$) for which $\omega_{v_{op}}^2(0) > 0$. We therefore see that (11) holds to order $(M_2/M_1)^{1/2}$ as $\mathbf{q} \rightarrow 0$. Since we are interested in the case $(M_2/M_1) \ll 1$, we define a small parameter $x = (M_2/M_1)^{1/2}$ and show that (11) holds for all \mathbf{q} , to order x .

To proceed, it is convenient to re-write (A.1) in the following form:

$$\begin{pmatrix} x^2 F^{11}(\mathbf{q}) & x F^{12}(\mathbf{q}) \\ x F^{21}(\mathbf{q}) & F^{22}(\mathbf{q}) \end{pmatrix} \begin{pmatrix} \mathbf{e}(\mathbf{q}|v1) \\ \mathbf{e}(\mathbf{q}|v2) \end{pmatrix} = \omega_v^2(\mathbf{q}) \begin{pmatrix} \mathbf{e}(\mathbf{q}|v1) \\ \mathbf{e}(\mathbf{q}|v2) \end{pmatrix} \quad (\text{A.4})$$

with $F^{ss'}(\mathbf{q})$ a 3×3 tensor defined by

$$F_{ij}^{ss'}(\mathbf{q}) = (M_2)^{-1} \sum_l \Phi_{ij}(l s; l' s') \exp\{i\mathbf{q} \cdot [\mathbf{x}(l) - \mathbf{x}(l')]\} \quad (\text{A.5})$$

and matrix multiplication on the left hand side of (A.4) is implied.

Expanding (A.4) we obtain:

$$\begin{aligned} x^2 F^{11}(\mathbf{q}) \mathbf{e}(\mathbf{q}|v1) + x F^{12}(\mathbf{q}) \mathbf{e}(\mathbf{q}|v2) &= \omega_v^2(\mathbf{q}) \mathbf{e}(\mathbf{q}|v1) \\ x F^{21}(\mathbf{q}) \mathbf{e}(\mathbf{q}|v1) + F^{22}(\mathbf{q}) \mathbf{e}(\mathbf{q}|v2) &= \omega_v^2(\mathbf{q}) \mathbf{e}(\mathbf{q}|v2). \end{aligned} \quad (\text{A.6})$$

Note that $F^{ss'}(\mathbf{q})$ is independent of x . We now expand all quantities in power series in x so that

$$\begin{aligned}\omega_v^2(\mathbf{q}) &= \omega_{v(0)}^2(\mathbf{q}) + x \omega_{v(1)}^2(\mathbf{q}) + \dots \\ e(\mathbf{q}|v_s) &= e^{(0)}(\mathbf{q}|v_s) + x e^{(1)}(\mathbf{q}|v_s) + \dots\end{aligned}\tag{A.7}$$

Substituting (A.7) into (A.6), and equating powers of x on the right and left hand sides, we obtain the zero-order equations

$$\omega_{v(0)}^2(\mathbf{q}) e^{(0)}(\mathbf{q}|v_1) = 0\tag{A.8}$$

$$\omega_{v(0)}^2(\mathbf{q}) e^{(0)}(\mathbf{q}|v_2) = F^{22}(\mathbf{q}) e^{(0)}(\mathbf{q}|v_2).\tag{A.9}$$

Using (A.8), we see that there are three solutions for which $\omega_{v_{ac}(0)}^2(\mathbf{q}) = 0$ and $e^{(0)}(\mathbf{q}|v_{ac}1) \neq 0$ which define the acoustic mode branches ($v = 1, 2, 3$). From (A.9), we see that for these modes, $e^{(0)}(\mathbf{q}|v_{ac}2) = 0$ is a solution. Similarly, from (A.9) we find three modes $\omega_{v_{op}(0)}^2(\mathbf{q})$, ($v = 4, 5, 6$), which are the eigenvalues of $F^{22}(\mathbf{q})$ and for which $e^{(0)}(\mathbf{q}|v_{op}2) \neq 0$. These modes have positive square frequencies (crystal stability), and are the three optic mode branches (note that $\omega_{v_{op}(0)}^2(\mathbf{q})$ can vanish only if $\det |F^{22}(\mathbf{q})| = 0$, which can only occur at isolated points). From (A.8), $e^{(0)}(\mathbf{q}|v_{op}1) = 0$ for these modes.

Using the above results, and in addition considering the equations of first and second order in x , the results may be summarized as follows

$$\begin{aligned}v = 1, 2, 3 \quad \omega_v^2(\mathbf{q}) &= 0 + O(x^2) \delta_{q,0} \\ \text{acoustic mode branches} \quad \frac{|e(\mathbf{q}|v_2)|}{|e(\mathbf{q}|v_1)|} &= O(x)\end{aligned}\tag{A.10}$$

$$\begin{aligned}v = 4, 5, 6 \quad \omega_v^2(\mathbf{q}) &= \text{eigenvalues of } F^{22}(\mathbf{q}) + O(x^2) \\ \text{optic mode branches} \quad \frac{|e(\mathbf{q}|v_1)|}{|e(\mathbf{q}|v_2)|} &= O(x).\end{aligned}\tag{A.11}$$

Therefore, we see that equation (11) is valid to $O[(M_2/M_1)^{1/2}]$, and in addition, (A.10) and (A.11) show the existence of the acoustic-optic mode frequency gap. Since the theory for the EPI involves terms of the form $|e(\mathbf{q}|v_s)|^2$, corrections to our results in equation (13) are $O[(M_2/M_1)]$, as stated in the text.

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